Distributions of environmental radionuclides in a marine core from the eastern continental shelf of Hainan Island, South China Sea and risk assessment

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Abstract
Several long half-life natural and anthropogenic radionuclides, including ^{137}Cs, ^{226}Ra, ^{232}Th, and ^{40}K, in a 45 cm-long sediment core collected from the east continental shelf of Hainan Island, South China Sea, have been analyzed. The results showed that mean activity of ^{137}Cs, ^{226}Ra, ^{232}Th, and ^{40}K were 2.03 ± 1.2, 20.4 ± 1.8, 45.7 ± 3.0, and 520.0 ± 20.0 Bq/kg, respectively. Radium equivalent activity and External hazard index imply a low radiation, and the study area could serve as a background reference value for environmental radionuclides in east continental shelf of Hainan Island, South China Sea.

Keywords South China Sea · Marine sediments · Radionuclides · Radiological risk assessment

Introduction

Environmental radionuclides are omnipresent in the Earth environment. Radionuclides with different half-lives may derive from various source materials, making them ideal tools to trace geochemical processes, examine rock ages, and construct the chronology of sediment profiles [1, 2]. However, radionuclides may pose some potential health hazards to human beings through continuous internal and external irradiation [3]. Natural and anthropogenic radionuclides can enter the marine environment through several pathways, such as the discharge of radioactive wastes produced by nuclear tests or nuclear plants, leakage associated with nuclear accidents, weathering and erosion of terrestrial rocks and subsequent transportation by water, wind and gravity, etc., through surface running and groundwater infiltrating. The ocean acts as a sink and reservoir of various terrestrial

materials and marine sediments, which have a high capacity to absorb some radioactive materials [4]. Thus, marine sediments can well record the sedimentary history of radionuclides in the environment [5]. Radionuclides in marine sediments could also play a positive role in chronological analysis, environmental radiological risk assessment, and establishment of environmental radiation safety standards [6–9].

Some areas on the southern coast of China are typical high background radiation areas [10]. In addition, 16 nuclear power plants are currently in regular use in China's coastal areas and more are under construction or proposed for consideration [11]. Terrestrial materials from high radiation background areas and nuclear ore processing by nuclear power plants may release natural and anthropogenic radionuclides [12, 13], which may ultimately enter the ocean.

In addition, based on recent geological survey, continental shelf areas surrounding the Hainan Island were found to be enriched in marine sand resources [14]. These resources are good engineering construction materials with high quality, and future exploitation are under consideration according to the planning of the Chinese government (https://en.zgss.org.cn/). The sampling site of this study is located within the planning exploitation area. Thus, radiological risk assessment of these sediments is of great significance. To date, time-series distributions of environmental radionuclides in marine sediments, and their radiological risk in coastal areas are largely unknown. One of our aims is to examine and

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compare overall radioactivity of the marine sediments in ancient and recent times, respectively.

To assess and monitor the environmental risk of radionuclides, radium equivalent radioactivity (Ra$_{eq}$), which incorporates long half-life radionuclides of ^{226}Ra, ^{232}Th and ^{40}K, was proposed by scientists [15–19]. It has been widely used to assess radiological risk of marine sediments. For example, Sam et al. [20] analyzed the distributions of natural and man-made radionuclides in the surface sediments of the Sudan and Suakin ports along the Sudan coast of the Red Sea, and identified the influence of organic matter on radionuclide distributions. By analyzing natural radionuclides in the surface sediments of the Vizag coast in southeastern India, Tripathi et al. [21] reported a low level of radioactive hazards. However, soil/beach sand samples in Kalpakkam, India, have much higher levels of long half-life radionuclides, resulting in higher Ra$_{eq}$ [22]. In addition, a few studies were performed on radionuclides in marine sediments from China. For instance, Liu et al. [23] measured the spatial distributions of radionuclides in the sediments of the Beibu Gulf, South China Sea and reported radioactivities decreasing with the offshore distance. Wang et al. [24] and Lin et al. [25] determined radionuclides in coastal wetlands and marine sediments of the Laizhou Bay, and discussed their potential environmental risks. The external hazard index (H$_{ex}$) is also a good marker for radiological risk [21, 26–28]. In terms of risk assessment of radionuclides in soil or sediments, most of the previous research focused on spatial distribution [29–33], whereas time-series analysis in environmental samples is extremely insufficient.

This study was designed to reconstruct the records of several environmental radionuclides in the sediments of the eastern continental shelf of Hainan Island, South China Sea, and to examine the long-term changes in potential radiological risk. Our study also aids for future management and exploitation of marine sand resources, and helpful for understanding radiological baseline of the study area.

Materials and methods

Sample collection

The South China Sea (SCS) is located in the Western Pacific warm pool area and is the largest semi-enclosed marginal sea in the region. It is one of the regions with the highest level of biodiversity in the world [34, 35]. Due to its unique geographical location, the SCS is very sensitive to environmental changes and thus an ideal area for studying global and regional environmental changes [36]. The Pearl River in China and, the Red River, and the Mekong River in Vietnam are the major rivers entering the SCS [34]. These rivers provide the main source materials for the sediments on the shelf area of the SCS. The study area is located on the eastern continental shelf of Hainan Island, northern SCS.

According to previous studies of [37, 38], terrestrial input of the Pearl River is the main sediment source in the northern shelf area of the SCS. Over the past decades, countries around the SCS have been experiencing the fastest economic development with more than 270 million people living in the coastal areas of the SCS. The Pearl River Delta and its city cluster are the most economically developed areas in China, with a large population and high density. To relieve the pressure on energy and resources, the Chinese government has constructed several nuclear power plants on the northern coast of the SCS, and more are under consideration (Fig. 1).

The sediment core analyzed in this study was collected during the "National Natural Science Foundation of China (NSFC) sponsored Open Scientific Expedition Cruise to the western SCS" in 2015 and was named QD2. Though we used one core in the present study, we believe it can generally reflect regional environmental changes. Sediments on the eastern and northern coasts of the South China Sea share a common source, i.e. the Pearl River [37]. The sedimentary environment is thus relatively simple. Similar material sources in this area probably record similar deposition history. Marine productivity record based on QD2 coupled with large scale climatic (temperature, monsoon, etc.) changes [39]. This also partly suggests that the sampling site may serve as a good record for a large area.

The water depth of the sampling site, approximately 20 km offshore, is 84.6 m (coordinates 18°54.00′N, 110°42.00′E; Fig. 1). The sampling equipment was supported by vessel "Experiment No. 3" (owned by the South China Sea Institute of Oceanology, Chinese Academy of Sciences). A box corer with a height of 60 cm was used as a sampler. After the sampler was retrieved from the seafloor to the deck, the seawater was siphoned out. A 7.5 cm diameter push core was then inserted into the box corer. The pipe was sealed with made-to-fit ends, and the core was 45 cm in length. Prior to analysis, it was brought back to the laboratory and stored at − 20 °C.

The core was opened in the laboratory and sectioned at intervals of 1 cm (45 samples in total). The core was photographed and its lithological characteristics were also recorded in detail. In general, the core could be divided into two parts from the top toward the bottom. The upper part of the core from 0 to 20 cm is composed of yellow–gray fine sand, while sediments at the depth of 20–45 cm are mainly gray–black silt with some tiny mollusk shells [39]. Chronology analysis also revealed that sample age increased with depth (see Results and discussion), and no evidence of bioturbation was observed during core slicing. We are thus confident that the sediments have been well-preserved and the core serves as a natural archive.

Fig. 1 Map showing the sampling site and some nuclear power plants (NPPs) on the coast (based on Ocean Data View to create; the red filled circle shows the location of QD2 (which means Qiong-Dong area of the South China Sea); ☢: the running nuclear power units; ☢: NPPs under consideration. The marked numbers in the figure represent the nuclear power plants: ① Ningde NPP; ② Fuqing NPP; ③ Zhangzhou NPP; ④ Taipingling NPP; ⑤ Daya Bay and Lingao NPP; ⑥ Taishan NPP; ⑦ Yangjiang NPP; ⑧ Fangchenggang NPP; ⑨ Changjiang NPP). (Color figure online)
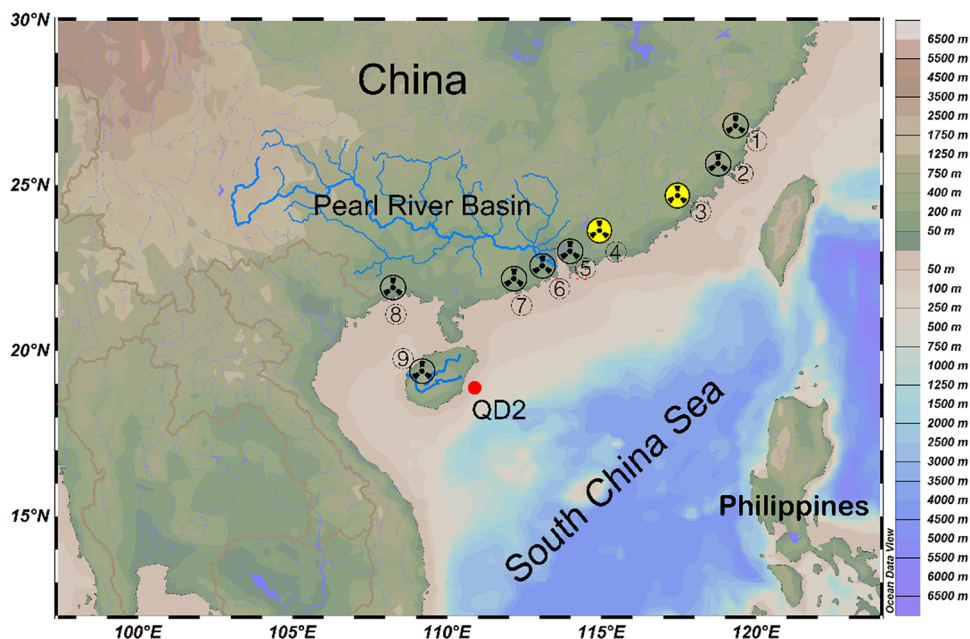
Analytical methods

Chronological and grain size analysis

The age-depth model of the core QD2 was constructed by both ^{210}Pb analysis of the surface sediments and radiocarbon (^{14}C) dating of foraminifera samples in the lower part sediments. Details of the chronological analysis were described in Ji et al. [39, 40]. Grain size composition of each sample was determined with a laser particle size analyzer after the samples were treated with excess H_2O_2 and HCl. The analyzer model is Malvern Mastersizer 2000 with a detection range of 0.02–2000 μm.

Radionuclide analysis

The radionuclides test is specified as follows: after freeze-drying, each sample was ground with an agate mortar and pestle to pass a 100-mesh sieve. Approximately 5–6 g of the sample was transferred to a standard centrifugal tube. The tube was then sealed with cyanoacrylate glue and stored for three weeks to reach the secular radioactive equilibrium between ^{226}Ra and its daughter isotopes. Sealing is very important for reaching equilibrium between ^{222}Rn and ^{226}Ra in the present study. Prior to experiment, we performed leak detection on the tubes we used. A centrifugal tube was sealed with cyanoacrylate glue and then soaked into a 500 mL beaker. No any bubble was observed during this process, implying that leakage of air was impossible. Measurements of the activity concentrations of ^{137}Cs, ^{226}Ra, ^{228}Ra and ^{40}K in Bq/kg (in dry weight) of the collected samples were determined using low-background high-purity germanium

(HPGe) gamma spectrometer. The counter has an energy resolution better than 2.3 keV at 1332 keV (Co-60). Several radionuclides, including ^{241}Am, ^{155}Eu, ^{57}Co, ^{54}Mn, ^{65}Zn and ^{60}Co (provided by National Institute of Metrology of China), were used to calibrate the spectrometer. The counting time for each sample was approximately 24 h. The detection limit for ^{137}Cs is 0.19 Bq/kg, and the limit for ^{226}Ra/^{228}Ra/^{40}K is 4.5 Bq/kg. ^{137}Cs was measured at the position of 661.7 keV. Radioactivity of ^{226}Ra was obtained by the weighted mean of its daughter isotopes ^{214}Pb at 295 keV and 351.9 keV and ^{214}Bi at 609.3 keV. For ^{228}Ra measurement, the weighted mean was taken from signals at the 338.3 keV, 911.2 keV, and 968.8 keV peaks of ^{228}Ac. The photopeak for ^{40}K was 1460.8 keV.

Results and discussion

Chronology and grain size compositions

According to the age-depth model constructed for QD2 by ^{210}Pb and ^{14}C tests, the ages of the QD2 samples increase with the depth without age inversion, implying good preservation of the samples. The age of the bottom sample of this core is 1852 years before present (106 AD), suggesting the core has a history of approximately 1900 years [39, 40] (Fig. 2).

Grain size compositions of sediment samples in QD2 were provided in Fig. 3. It tells that the sediments mainly consist of coarse components, i.e. sand and silt. In contrast, the content of clay is low. Additionally, the concentrations of typical land-origin element Aluminum (Al)
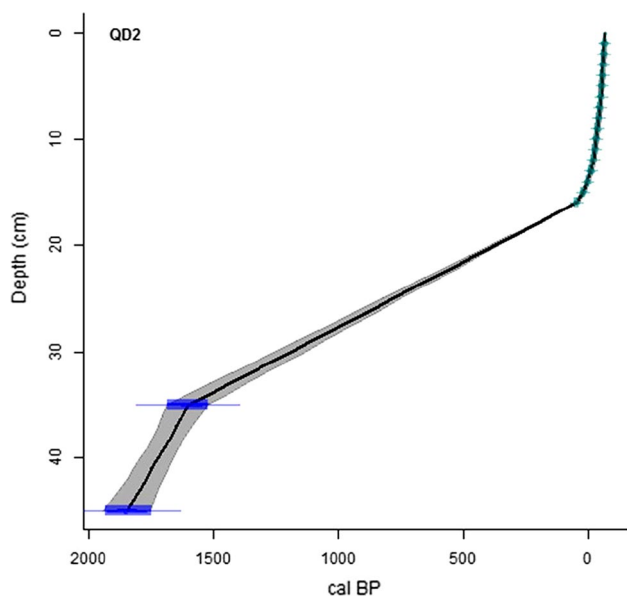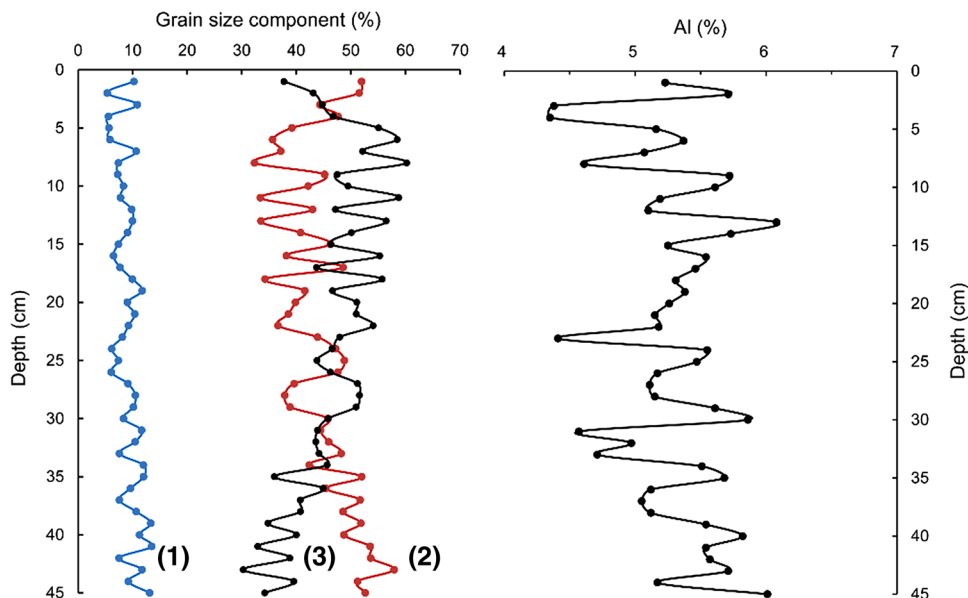
Fig. 2 Age-depth model for the profile QD2 based on ^{210}Pb and ^{14}C dating (Reprinted from [40] with permission from Wiley)

in this core are as high as ~ 4–6% [39]. This implies terrestrial input is the dominant source of these sediments. This is in agreement with an earlier study which reported that sediments from this study area were derived from the Pearl River [37].

Based on statistics, both sand and silt have relatively small coefficients of variation (CV) (~ 15%). Together with the relatively uniform lithology, the results suggest that the sediments in the study site could be in a relatively stable sedimentary environment, and the source material has not changed significantly.
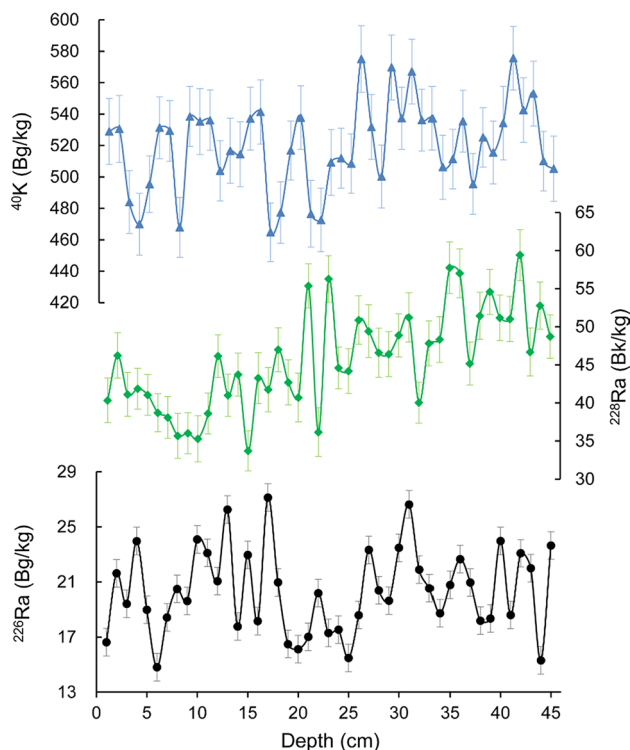
Distribution of nuclides in the sedimentary profile

Among the radionuclides that we analyzed, ^{137}Cs has the lowest radioactivity. It was found in the sediments above depth of 5 cm, and the radioactivity concentrations for these five samples are 4.0, 2.2, 1.8, 0.7, and 1.4 Bq/kg, respectively. The ^{137}Cs signal in the upper most layer could be a record of Fukushima Daiichi nuclear disaster in 2011. [137]Cs shows a small peak at the depth of 5 cm, and this might be measured from the Chernobyl nuclear accident. The distribution of ^{137}Cs in core QD2 also suggests a relatively low sedimentation rate. The missing of ^{137}Cs in deep layers could be explained by its natural decay, as it has a relatively "short" half-life in comparison with [226]Ra, [228]Ra and [40]K. The anthropogenic ^{137}Cs data also imply an insignificant impact of human activity on the sediments.

The activity concentrations of long-lived natural radionuclides [226]Ra, [228]Ra, and [40]K in QD2 sediment core are given in Fig. 4.

Figure 4 shows that the radioactivity of [226]Ra in QD2 ranges from 14.8 to 27.1 Bq/kg with an average of 20.4 ± 1.8 Bq/kg (n = 45). The minimum value appears at the depth of 6 cm and the maximum appears at 17 cm. The radioactivity of [226]Ra between 18 and 30 cm is slightly lower, but its overall trend is not evident.

The radioactivity of [226]Ra in QD2 is slightly lower than the worldwide average concentration of 35 Bq/kg [41] and significantly lower than most of the reported radioactivities of soil and sediment samples in China (Table 1). For example, [226]Ra radioactivities in soil samples of the Pearl River Delta and surface sediments from Nansha are as high as 135.9 Bq/kg and 80.6 Bq/kg, respectively [42, 43]. The Huangmao Sea-Guanghai Bay and adjacent sea areas and

Fig. 3 Down core distributions of grain components [(1) clay, (2) silt and (3) sand] and Al (the data of Al are cited from reference [39])

Fig. 4 Radioactivity-versus-depth profiles of ^{226}Ra, ^{228}Ra, and ^{40}K in QD2 core with depth

the Yangjiang nuclear power sea area also have much higher activity concentrations of ^{226}Ra [10, 44]. This large variability of ^{226}Ra could be partly attributed to the type of soil/sediments, soil parent material, landform and radiochemistry in specific areas. For instance, the Pearl River Delta watershed is an area with high radiation background [45, 46], leading to a relatively high level of ^{226}Ra in its soil sediments. In terms of the absolute values, the radioactivity of ^{226}Ra in QD2 is approximately half of the average radioactivity of ^{226}Ra in Chinese soil, i.e. 37.6 Bq/kg [47].

Compared to ^{226}Ra, ^{228}Ra in the QD2 sediments has higher radioactivity ranging from the minimum of 33.7 Bq/kg (at 15 cm) to the maximum of 59.4 Bq/kg (at 42 cm), with an average of 45.7 ± 3.0 Bq/kg (n = 45). Throughout the entire profile, the radioactivity of ^{228}Ra in the upper part (0–20 cm) of the core is lower than that of the lower sediments (Fig. 4). From a global perspective, the ^{228}Ra radioactivities in the QD2 core are higher than the international average of 30 Bq/kg [41].

Among the analyzed radionuclides, ^{40}K in QD2 core shows relatively higher radioactivity level (Fig. 4) between 464.8 and 575.6 Bq/kg, with an average of 520.0 ± 20.0 Bq/kg. The radioactivity of ^{40}K in QD2 core was higher than those in some previously investigated coastal areas of China, e.g. the Beibu Gulf, Bailong Peninsula, Pearl River Estuary and Nansha sea area (253–511.5 Bq/kg) [48–52]. Conversely, the radioactivity of ^{40}K in QD2 core was overall

Table 1 The Ra$_{eq}$ values in marine and estuary sediments

Area	Radioactivity (Bq/kg)			Ra$_{eq}$ (Bq/kg)	References
	^{226}Ra	^{228}Ra	^{40}K		
South China Sea Coral Reef	3.3	5.1	24.4	12.5	[25]
Beibu Gulf*	22.2	34.4	253.0	90.9	[48]
Pearl River Estuary, Dapeng Bay and Daya Bay	27.9	36.5	456.2	115.2	[50]
Shenzhen coastal areas	26.5	43.2	364.2	116.3	[51]
The adjacent areas to Bailong Peninsula, Beibu Gulf*	32.4	46.1	355.0	125.7	[49]
Continental shelf areas of Hainan Island	20.4	45.7	520.0	125.7	This study
Yangtze River Estuary	24.3	40.9	628.0	131.1	[71]
Northeast of South China Sea	27.7	44.9	538.0	133.3	[23]
Yangtze River Estuary and adjacent areas*	18.5	48.5	684.0	140.5	[72]
Hong Kong sea areas	32.5	48.1	625.0	149.4	[73]
Changyi Coastal Wetland/Laizhou Bay	28.6	57.9	542.0	153.1	[15]
Tianwan Sea Area, Lianyungang	25.2	46.7	899.0	161.2	[56]
Jiaozhou Bay	26.5	40.3	688.0	164.4	[53, 74]
Yangjiang Nuclear Power Sea Area	35.5	57.1	621.0	165.0	[10]
Huangmao Sea-Guanghai Bay and adjacent sea areas	36.6	61.3	571.0	168.2	[44]
Xinghua Bay, Fujian	24.8	63.7	734.0	172.4	[55]
Intertidal zone in Xiamen sea areas	32.4	69.3	692.0	184.8	[54]
Nansha sea area	80.6	45.5	511.5	185.0	[52]
Pearl River Delta	135.9	196.9	670.2	469.1	[42]

Ra$_{eq}$ values in areas marked with asterisks were given in the cited papers. Ra$_{eq}$ in areas without asterisks were calculated by nuclide activity values provided in the respective paper(s)

lower than those in sediments from the northeastern of SCS, Laizhou Bay, Xinghua Bay, Taiwan sea and other investigated areas (538–899 Bq/kg) [10, 43, 53–56]. In general, the average radioactivity of ^{40}K in QD2 core is slightly lower than the average radioactivity of ^{40}K in China (584.0 Bq/kg), but higher than the global average of 400 Bq/kg. To further assess the overall long-term environmental risk of radionuclides in the study area, we calculated the Ra$_{eq}$ index based on the measured radioactivity concentrations of ^{226}Ra, ^{228}Ra and ^{40}K.

Radiological risk assessment of radionuclides in core QD2

Individual radionuclides may not reflect the overall long-term risk of the bulk sediments, and an integrated marker is thus required. Hazard indexes could be good indicators that reflect general radioactivity of environment samples. Such indexes are thus employed to investigate changes in radiation of marine sediments on millennial scales. The utilization of hazard indexes to examine marine sediments was generally acknowledged by scientific community, and the indexes have been applied to some ocean areas, including the Bay of Algeciras [57], shore of Vizag [21] and the southern South China Sea [58]. As stated above, the core dates back approximately 1900 years. Together with the established chronology, we used radium equivalent activity and external hazard index to evaluate radiological risk over the last ~1900 years.

Radium equivalent activity (Ra$_{eq}$)

In order to compare the comprehensive radiological effects of the main natural radionuclides in the continental shelf of eastern Hainan Island, Ra$_{eq}$ was used to evaluate the radioactivity of each sample in QD2 sediment core, which is widely used in radioactive risk assessment [59–61]. The hazard index is calculated based on the activity of natural long half-life radionuclides ^{226}Ra, ^{232}Th and ^{40}K, mathematically defined as follows [62–65]:

$$Ra_{eq} = A_{Ra} + 1.43 \times A_{Th} + 0.077 \times A_K \qquad (1)$$

where A_{Ra}, A_{Th}, A_K are the activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K in the sediments, respectively. It is generally accepted that the radioactivity of ^{232}Th is equal to its decay daughter ^{228}Ra due to secular equilibrium [26, 28, 63, 66]. Thus, ^{228}Ra is frequently used as a substitution for ^{232}Th in the calculation of Ra$_{eq}$. Based on chronology and activity data, we have constructed a 1900-year record of Ra$_{eq}$ for the QD2 core (Fig. 5), which is the first long-term, i.e. millennial-scale, Ra$_{eq}$ record.

Figure 5 shows the minimum value of Ra$_{eq}$ is 107.5 Bq/kg at the year of 1989 AD, and the maximum (149.8 Bq/
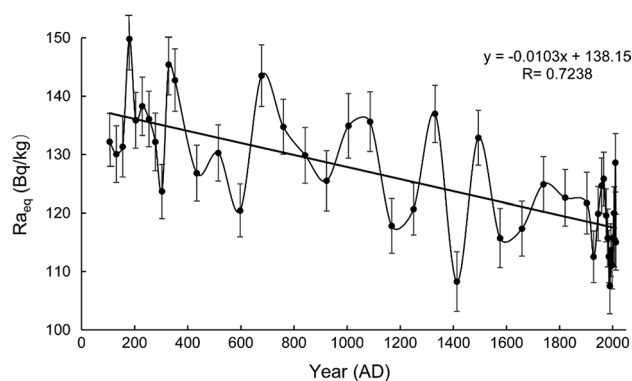
Fig. 5 Ra$_{eq}$ values for sediments in the core QD2 over the past 1900 years

kg) appears at 180 AD. The present-day Ra$_{eq}$ of 115.0 Bq/kg is approximately 77% of the maximum. The average Ra$_{eq}$ over the past 1900 years is 125.7 Bq/kg ± 10.4 (n = 45). In general, Ra$_{eq}$ of 370 Bq/kg is widely accepted by the scientific community as a safety threshold [34, 67–70]. In comparison with this recommendation value, the Ra$_{eq}$ of sediments from the eastern continental shelf of the Hainan Island, SCS, is significantly lower. This suggests that the radiological hazard in the study area is low. It is also noted that Ra$_{eq}$ shows a decreasing trend in the past 1900 years (Fig. 5) and is currently at a relatively low level, which is only 31% of the safety threshold.

We compared the Ra$_{eq}$ values of marine and estuary sediments in some other areas of China by compiling the published Ra$_{eq}$ data (Table 1). According to this table, sediment Ra$_{eq}$ values in other regions of China are generally low, ranging from 12.5 to 469.0 Bq/kg. The overall radioactivity level of the QD2 samples is similar to that of marine sediments in some coastal areas of China (such as the sediments adjacent to the Bailong Peninsula in Beibu Gulf and the surface sediments in the Yangtze River estuary) but lower than the average radioactivity of the Chinese soils (165.7 Bq/kg)[42].

Due to the widespread granite and acidic volcanic rocks around the Pearl River Delta, northern coast of the South China Sea is a high radiation background area and geological samples there contain high levels of uranium and thorium, and its Ra$_{eq}$ (469.1 Bq/kg) exceeds the safety threshold of 370 Bq/kg (Table 1) [10, 42, 73, 74]. All the other investigated areas in China, including the coast area in this study, have Ra$_{eq}$ values below the threshold, further attesting to a low radiological risk. Therefore, the Ra$_{eq}$ data obtained in this study can serve as the background value of radioactivity in the study area. The average value can be further used as an environmental background to guide environmental management and decision-making concerning environmental radioactivity.

We also investigated the radioactivities of radionuclides in samples around the globe. Ra_{eq} data from different regions of the world are presented in Fig. 6 [47, 75–78]. Figure 6 shows most studies on soil/sediment radioactivity are from Europe [20, 41, 79], East Asia and South Asia [22, 56, 74, 80], with a few related studies from West Asia [81–83]. Comparatively, studies of Ra_{eq} from Africa, Australia and America are scarce [26, 84, 85].

The average radioactivity of QD2 samples is (125.7 Bq/kg) slightly higher than that of the world average of 108.7 Bq/kg [41]. Available global data show the radioactivities of sediments and soils from Europe are relatively low [41, 79]. The European average Ra_{eq} value is very close to that of QD2 samples in this study (125.7 Bq/kg), and the lowest radioactivity (43.1 Bq/kg) is from the Black Sea coast of Turkey [86]. Among all the data we collected, the maximum Ra_{eq} is from Kalpakkam, India [22], which is as high as 644.1 Bq/kg, about four times greater than sediments in our study. From a global perspective, the average radioactivity level in sediments from the eastern continental shelf of Hainan Island, SCS, is at a medium level and below the safety threshold. Therefore, the study area is at low radiological risk.

External hazard index (H$_{ex}$)

The external hazard index (H$_{ex}$) is also a good indicator of radiological risk. The sediments in the marine environment mainly come from the nearby land continent. Thus, H$_{ex}$ can be used to estimate the hazard of natural gamma radiation of the marine sediments and the terrestrial mainland [21, 58, 87]. The calculation formula is as follows [21]:

$$H_{ex} = A_{Ra}/370 + A_{Th}/259 + A_{K}/4810 \qquad (2)$$

The results revealed that H$_{ex}$ values of QD2 core samples ranged from 0.29 to 0.40, with an average of 0.34, less than the safety threshold value 1 [26, 65, 88, 89]. In comparison with published some H$_{ex}$ values of marine sediments (e.g. Southeast India, Egyptian Red Sea Coast, Southern Italy and Upper Gulf of Thailand etc.), our values are low. This also suggests that the risk of external radiation hazards is very small.

Based on Ra_{eq} and H$_{ex}$, together with the pretty low level of ^{137}Cs, it is concluded that the delivery of land-based materials to the sediments and nuclear power plant operations (including nuclear ore processing) nowadays in China's coastal areas do not cause great radiation hazards.

Conclusions

A sediment core QD2 was collected from the eastern continental shelf of Hainan Island, SCS. Several radionuclides, including ^{137}Cs, ^{226}Ra, ^{228}Ra and ^{40}K were determined. The activities of ^{137}Cs are very low, suggesting a minor impact of anthropogenic impact. Average activities of ^{226}Ra, ^{228}Ra and ^{40}K are 20.4 ± 1.8 Bg/kg, 45.7 ± 3.0 Bg/kg, and 520.0 ± 20.0 Bq/kg. In combination with chronological analysis, the time-series distributions of ^{226}Ra, ^{228}Ra, ^{40}K in the sediments of the core were reconstructed. Radium
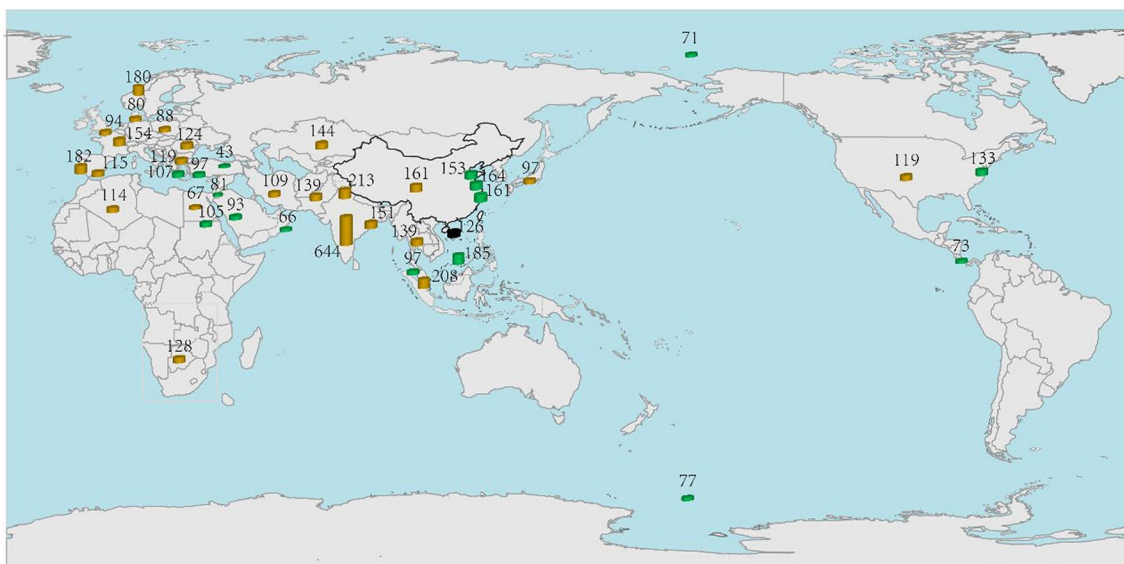
Fig. 6 Global distributions of soil/sediment Ra_{eq} values (black bar denotes marine sediments in this study; green and yellow markers represent sediment and soil samples, respectively). (Color figure online)

equivalent activity (Ra_{eq}) and external hazard index (H_{ex}) were employed to evaluate the radiation risk. The mean value of Ra_{eq} and H_{ex} are 125.7 Bq/kg and 0.34, respectively. The Ra_{eq} value of QD2 is generally low with an average of 125.7 Bq/kg, much smaller than the Ra_{eq} safety threshold (370 Bq/kg). Thus, the environmental risk associated with radionuclides around the study area should be low. In combination with chronological analysis, a 1900-year record of Ra_{eq} was reconstructed, and the record shows a decreasing trend in Ra_{eq}. H_{ex} average is less than the safety threshold 1. Both Ra_{eq} and H_{ex} suggest a low radiological risk. Together with low level of ^{137}Cs, it is concluded that the environmental risk associated with radionuclides around the study area should be low. The eastern continental shelf area of Hainan Island, SCS, could serve as a natural background in environmental radiation studies. The results of this study provide reference values for the assessment of marine radiation safety in coastal areas and monitoring of radionuclide pollution.

Author contribution All authors contributed to the study conception and design. Material preparation, data collection and analysis were performed by Fengmei Wang, Chao Ji and Lingling Jiang. The first draft of the manuscript was written by Fengmei Wang and all authors commented on previous versions of the manuscript. All authors read and approved the final manuscript.

Declarations

Conflict of interest The authors have no conflicts of interest to declare that are relevant to the content of this article.

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