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Baseline

Natural radioactivity assessment of surface sediments in the Yangtze Estuary

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ABSTRACT

The activities of the natural radionuclides (^{238}U, ^{232}Th, ^{226}Ra and ^{40}K) of the surface sediments in the Yangtze Estuary were determined and used to evaluate radiation hazards in the study area. The of activities of ^{238}U, ^{232}Th, ^{226}Ra and ^{40}K ranges from 14.1 to 62.3, 26.1 to 71.9, 13.7 to 52.3, and 392 to 898 Bq kg^{-1}, respectively, which were comparable to values of other regions in China. The activities of ^{232}Th, ^{40}K and ^{226}Ra were clearly different from the global recommended values. The radium equivalent activity was less than the recommended limit of 370 Bq kg^{-1}; therefore, the sediment in this area can be safely used for reclamation. The external hazard index values were less than one. The average absorbed gamma dose rate and annual effective dose equivalent values were slightly greater than the world average value. $^{226}Ra/^{238}U$ and $^{232}Th/^{238}U$ ratios could potentially be applied for tracing sediment source.

Naturally occurring radionuclides (e.g., thorium, uranium and their daughter nuclides) are widespread in the earth surface systems, such as the lithosphere, biosphere, and pedosphere (Freitas and Alencar, 2004; Abbady et al., 2006; González-Fernández et al., 2012; Qureshi et al., 2014; Ramasamy et al., 2014; Yasmin et al., 2014; Huang et al., 2015). Terrestrial gamma radiation from natural radioactivity occupies 85% of the total global annual average ionizing radiation (UNSCEAR, 2001). Long-term exposure to radioactivity and inhalation of radionuclides could cause many health problems, such as acute leukopenia, anemia, leukemia, necrosis of the mouth, tooth fracture and cataracts as well as lung, pancreatic liver, hepatic, bone and kidney cancers (Taskin et al., 2009; Qureshi et al., 2014). Human exposure to ionizing radiation is an important scientific subject that attracts sustained public attention. Among the various geological formations, sediment plays a predominant role in accumulating and transporting contaminants within a geographic area (Ramasamy et al., 2014). The study of natural radioactivity in sediments can provide useful information not only about sources, transport mechanisms and the environmental fate of radionuclides in aquatic environments (Huang et al., 2011, 2013; Wang et al., 2016) but also for the assessment of radiological risks and the control of radioactivity (Seddeek et al., 2005; Akram et al., 2006; Lu et al., 2008).

To estimate the natural radiation dose for different environments, previous research has been conducted on natural radioactivity levels in the sediments of rivers, lakes and coasts all over the world (Tsabaris et al., 2007; Śleziak et al., 2010; Agbalagba and Onoja, 2011; Eroğlu and Kabadayi, 2013; Tripathi et al., 2013; Ramasamy et al., 2014; Isinkaye and Emelue, 2015; Kritsananuwat et al., 2015). However, research concerning the natural radioactivity levels of estuarine sediment is limited. Natural radioactive elements present in sediment are primordial radionuclides from the ^{238}U series, ^{232}Th series and ^{40}K, although radionuclides from the ^{235}U series, cosmogenic nuclides and others can also contribute to the natural radiation dose. The objective of this paper is to observe the distribution of ^{238}U, ^{232}Th, ^{226}Ra and ^{40}K activities in the surface sediment of the Yangtze Estuary and to evaluate the radiological hazards in this area by calculating the radium equivalent activity (Ra_{eq}), absorbed gamma dose rate (D), annual effective dose equivalent (AEDE), and external hazard index (H_{ex}).

The Yangtze River is 6300 km long, drains a 1.96×10^6 km^2 area of China and is ranked third in the world in terms of flow volume (Beardsley et al., 1985). The Yangtze Estuary is a tide-dominated depositional basin. Shanghai, the most populous city in China, is located on the southern edge of the mouth of the Yangtze River. With rapid economic development in Shanghai and other cities in the river's drainage area, nutrient loading from the Yangtze River has increased dramatically, and these nutrients have been transported to the East China Sea over the two past decades (Gao and Song, 2005). As an international port city, Shanghai attracts large numbers of visitors each year from all over the world with its architecturally distinct buildings and beautiful nighttime views. Dozens of reclamation projects have taken place on Chongming Island over the 5 past decades. In particular, three large-scale reclamations performed in 1990, 1992 and 1998 increased the land area from ~6 to 12 million square kilometers and extended the seawall 10 km toward the sea (Yu and Bin, 2006). Sediment in the Yangtze Estuary is viewed as a potential resource for reclamations on Chongming Island and raw material for construction. The Yangtze Estuary is also the source of water for Shanghai and Jiangsu Province. Therefore, it is vital to evaluate the natural radioactivity level and radiological

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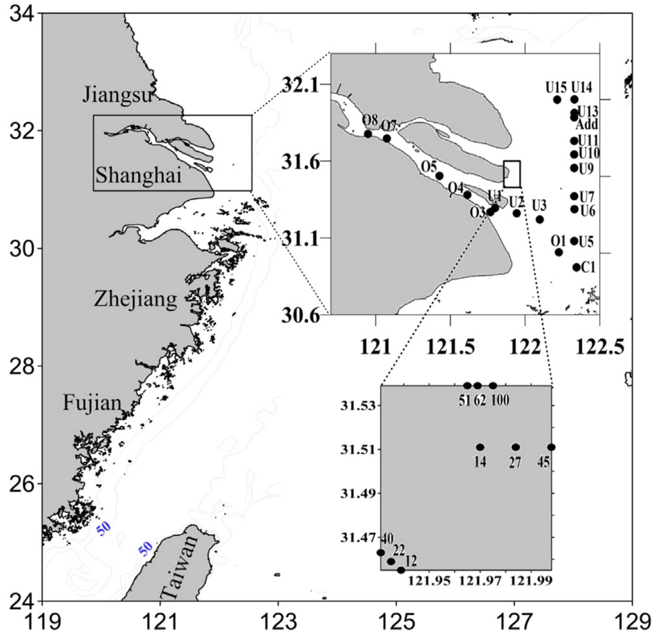

Fig. 1. Study area and sampling site locations.

hazards of the sediment in the Yangtze Estuary. Such an evaluation can contribute to the natural radioactivity level database of sediments in rivers and coastal areas of China and provide information for local radiation protection.

Twenty-nine surface sediment samples were collected using a box core and subsampled with a 10 cm (diameter) plexiglass core tube. The samples were taken on December 14, 2011, July 11–13, 2012 and August 14–15, 2013 (Fig. 1b). Six samples collected in the inner Yangtze Estuary, fourteen were collected outside of the Yangtze Estuary and nine were collected in the tidal flat on Chongming Island. The surface sediment was collected from the upper 1 cm of the surface using a stainless steel knife. The samples were put in resealable plastic bags and stored at 4 °C for future laboratory analyses. An aliquot of these samples was oven dried at 60 °C for several days and crushed. The dried sample was homogeneously pulverized, weighed and sealed in a plastic box (70 mm diameter × 70 mm height) for at least three weeks before counting to establish a secular equilibrium between ^{226}Ra and the daughter products of ^{222}Rn.

The activity of natural radionuclides ^{238}U ^{232}Th, ^{226}Ra, and ^{40}K in the samples was determined using the HPGe γ-ray detector (Canberra Be3830) with 35% relative counting efficiency and an energy resolution of 1.8 keV (at 1332 keV). The detector has multi-layer shielding, an ultra-low cryostat and no peak background in the isotopes of interest. All sediment samples measurements were taken at least 2 years after collection to ensure that the parent nuclides were in secular equilibrium with their daughter nuclides. The activity of ^{238}U was determined using the gamma line at 63.3 keV (4.84%) for ^{234}Th. ^{232}Th activity was analyzed using the mean activity of ^{228}Ra (^{228}Ac) and ^{228}Th. The activity of ^{228}Ra was determined using the gamma line at 338.3 keV (11.4%) and 911.2 keV (27.7%) for ^{228}Ac. ^{228}Th activity was determined by 238.6 keV (100%) and 583.2 keV (85.2%) gamma rays emitted from ^{212}Pb and ^{208}Tl. ^{226}Ra activity was determined using the gamma line at 351.9 keV (37.6%) for ^{214}Pb and 609.3 keV (46.1%) for ^{214}Bi. ^{40}K activity was measured directly through its gamma ray energy peak of 1460.8 keV (10.7%). The efficiency calibration of the detector systems was conducted using both Laboratory Sourceless Calibration Software (LabSOCS) and standard samples (GBW04127) to ensure the reliability of the QA/QC method. The detection limit of ^{232}Th, ^{226}Ra and ^{40}K is 2.0, 1.3 and 9.6 Bq kg^{-1}, respectively, for a counting time of 86,400 s.

The activities of ^{238}U, ^{232}Th, ^{226}Ra and ^{40}K in the sediment samples from the Yangtze Estuary are listed in Table. 1, ranging from 14.1 to 62.3 (mean: 32.8 ± 10.6 Bq kg^{-1}), from 26.1 to 71.9 (mean: 40.9 ±

Table 1
Activities (Bq kg^{-1}) of ^{238}U, ^{232}Th, ^{226}Ra and ^{40}K in the sediment from the Yangtze Estuary.

Sampling location	Sampling time	Longitude	Latitude	^{238}U (Bq kg^{-1})	^{232}Th (Bq kg^{-1})	^{226}Ra (Bq kg^{-1})	^{40}K (Bq kg^{-1})
14	2011/12/14	121.970	31.511	38.1 ± 11.3	41.1 ± 2.6	27.0 ± 1.2	559 ± 24
27	2011/12/14	121.984	31.511	29.8 ± 10.9	48.1 ± 3.2	25.6 ± 1.4	570 ± 28
45	2011/12/14	121.998	31.511	30.3 ± 9.5	41.4 ± 2.5	27.1 ± 1.2	392 ± 18
40	2011/12/14	121.931	31.463	46.3 ± 13.8	44.7 ± 2.9	29.0 ± 1.4	546 ± 26
22	2011/12/14	121.935	31.459	62.3 ± 17.6	71.9 ± 4.3	52.3 ± 2.0	376 ± 18
12	2011/12/14	121.939	31.455	30.8 ± 9.0	34.9 ± 2.2	25.4 ± 1.1	451 ± 20
51	2011/12/14	121.965	31.539	39.3 ± 13.0	50.0 ± 3.2	30.4 ± 1.5	585 ± 28.2
62	2011/12/14	121.969	31.539	51.0 ± 15.1	53.7 ± 3.3	32.1 ± 1.5	590 ± 27
100	2011/12/14	121.975	31.539	35.4 ± 11.5	46.0 ± 2.9	29.8 ± 1.4	592 ± 27
U1	2012/7/11	120.799	31.295	28.3 ± 2.7	41.5 ± 1.9	25.9 ± 1.4	525 ± 13
U2	2012/7/12	121.944	31.261	24.5 ± 3.2	46.5 ± 2.0	22.7 ± 0.9	778 ± 17
U3	2012/7/12	122.099	31.221	30.2 ± 2.8	40.8 ± 1.7	22.3 ± 1.3	679 ± 14
U5	2012/7/12	122.330	31.080	34.1 ± 2.9	45.8 ± 2.2	22.5 ± 1.6	794 ± 19
U6	2012/7/12	122.330	31.288	14.1 ± 2.1	26.1 ± 1.8	13.7 ± 0.3	482 ± 12
U7	2012/7/12	122.329	31.371	34.4 ± 2.7	34.5 ± 2.0	24.0 ± 1.6	702 ± 18
U9	2012/7/12	122.329	31.554	27.0 ± 3.4	28.8 ± 1.8	17.3 ± 1.3	665 ± 14
U10	2012/7/12	122.328	31.643	40.3 ± 3.7	42.9 ± 1.6	22.4 ± 1.2	798 ± 13
U11	2012/7/12	122.329	31.731	32.5 ± 2.3	42.7 ± 2.1	21.2 ± 1.5	846 ± 19
U13	2012/7/12	122.330	31.913	29.5 ± 3.0	37.2 ± 1.6	22.3 ± 1.2	605 ± 13
U14	2012/7/13	122.330	32.000	32.8 ± 3.4	34.6 ± 1.9	18.2 ± 1.4	563 ± 14
U15	2012/7/13	122.215	31.999	21.5 ± 3.2	36.3 ± 1.9	19.0 ± 1.4	605 ± 14
Add	2012/7/13	122.330	31.883	16.3 ± 2.9	30.1 ± 1.5	14.8 ± 0.7	559 ± 11
O3	2013/8/14	121.768	31.268	43.0 ± 6.3	38.5 ± 1.3	30.6 ± 0.8	650 ± 9
O4	2013/8/14	121.613	31.380	19.3 ± 7.9	28.3 ± 1.6	16.9 ± 1.1	733 ± 12
O5	2013/8/14	121.427	31.504	31.8 ± 7.4	45.1 ± 1.5	27.0 ± 1.1	592 ± 10
O7	2013/8/14	121.075	31.747	16.8 ± 8.6	31.3 ± 1.7	16.3 ± 1.2	640 ± 13
O8	2013/8/14	120.948	31.776	38.1 ± 11.6	55.3 ± 2.3	31.8 ± 1.6	818 ± 18
C1	2013/8/15	122.344	30.909	30.9 ± 11.6	42.9 ± 2.5	22.0 ± 1.6	840 ± 18
O1	2013/8/15	122.226	31.006	43.1 ± 10.2	41.6 ± 2.1	24.1 ± 1.4	898 ± 16
Range				14.1–62.3	26.1–71.9	13.7–52.3	392–898
Mean				32.8 ± 10.6	40.9 ± 9.4	24.3 ± 7.4	628 ± 135