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Radioactivity concentrations and their radiological significance in sediments of the Tema Harbour (Greater Accra, Ghana)

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ABSTRACT

Studies on environmental radioactivity in tropical Africa are scarce. Therefore, a baseline study of natural (^{238}U, ^{210}Pb, ^{226}Ra, ^{232}Th, ^{228}Ra, ^{228}Th, ^{40}K) and anthropogenic (^{137}Cs) radionuclides was carried out on Tema Harbour (Greater Accra, Ghana) surface sediments and on their radiological significance. Grab surface sediment samples were collected from 21 stations within the Tema Harbour and their radioactivity concentrations measured by gamma spectrometry. The mean sediment radioactivity concentrations (Bq kg^{-1} dw) were 34 for ^{238}U, 210 for ^{210}Pb, 14 for ^{226}Ra, 30 for ^{232}Th, 29 for ^{228}Ra, 31 for ^{228}Th, 320 for ^{40}K, and 1.5 for ^{137}Cs. Large ^{238}U/^{226}Ra disequilibria were observed in the harbour sediments and a complex dynamics of several mixed sources of sediments within the Tema Harbour can be inferred from the spatial variations in the radioactivity concentrations. The estimated total absorbed dose rate in air (D), radium equivalent activity (Ra_{eq}), external hazard index (H_{ex}), annual gonadal dose equivalent (AGDE) and annual effective dose equivalent (AEDE) indicated no significant radiological risks from the sediment radioactivity concentrations. Application of the Environmental Risk from Ionising Contaminants Assessment and Management tool (ERICA) confirmed that the potential dose rates to biota from the sediment radioactivity concentrations are unlikely to pose appreciable ecological risks. The radioactivity levels are compared with levels reported in sediments from other coastal areas of the world.

1. Introduction

Radionuclides constitute an important source of ionising radiation exposure to human and non-human populations (Kam & Bozkurt, 2007; UNSCEAR, 2000), which can cause harmful biological effects such as DNA damage and cancer (Little, 2003; Ravanat et al., 2014; Schmid & Schrader, 2007). Radionuclides such as ^{238}U, ^{210}Pb, ^{226}Ra, ^{232}Th, ^{228}Ra, ^{228}Th and ^{40}K are widely distributed in the environment as a result of their natural occurrence in the Earth's crust or the atmosphere. The human population worldwide receives an average annual radiation dose of 2.4 mSv.y^{-1}, about 80% of which comes from naturally-occurring radionuclides, the remaining part is largely due to artificial sources of which fallout radionuclides account for only 0.4% (UNSCEAR, 2000). Fallout radionuclides such as ^{90}Sr, ^{137}Cs and $^{239+240}$Pu are derived mainly from global nuclear tests conducted between the mid 1940s and the 1980s, as well as from nuclear accidents (Livingston & Povinec, 2000). In addition to their potential ionising effects, radionuclides may be toxic and can undergo bioconcentration and bioaccumulation (Hassona, Sam, Osman, Sirelkhatim, & LaRosa, 2008; Sirelkhatim, Sam, & Hassona, 2008) and adversely impact human and ecosystem health. Assessment of radioactivity in the environment is useful for the protection of human health and the environment from the harmful effects of ionising radiation, and is therefore of great interest (Ulanovsky, Pröhl, & Gómez-Ros, 2008). Most naturally-occurring and fallout radionuclides can be detected and measured at extremely low concentrations; this makes them excellent tracers for many environmental processes as well as

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unique dating tools. Thus, they have found a wide range of applications in environmental studies such as isotope hydrogeology (Divine & McDonnell, 2005), water masses circulation (Broecker, 1982), sediment dating and sedimentation (Hernández, 2016; Mabit et al., 2014; Mahu et al., 2016).

Although the West African coastal environment lacks the presence of nuclear industries, it may be impacted by contaminated areas as a result of ocean and atmospheric dispersal and redistribution. Furthermore, anthropogenic activities in the West African coastal environment such as shipping, offshore oil and gas exploration and production, mining, industrialisation, urbanisation, and agricultural production can potentially add to measured levels of radionuclides in the environment (Al-Trabulsy, Khater, & Habbani, 2011; El Mamoney & Khater, 2004; Nyarko et al., 2011). In the coastal environment, harbours may be particularly susceptible to anthropogenic influences and their sediments can act as sinks for radionuclides (Sugandhi, Joshi, & Ravi, 2014). Harbours have hence been areas of interest when investigating radionuclide contamination in the coastal environment (Akram et al., 2006; Kumar et al., 2013; Papaefthymiou, Papatheodorou, Moustakli, Christodoulou, & Geraga, 2007; Sam, ElGanawi, Ahamed, & ElKhangi, 1998; Sugandhi et al., 2014). Sediment contamination by radionuclides of the ^{238}U and ^{232}Th decay-series and ^{40}K is of particular interest from radiological point of view, as they can form the basis of radiological assessments for the human population. The Environmental Risk from Ionising Contaminants Assessment and Management tool (ERICA) developed by the European Commission provides an integrated approach to the assessment and management of environmental risks from ionising radiation (Beresford et al., 2007) and can be applied to assess the potential ecological impact of radionuclide-contaminated environments.

Currently, there is not much information on radioactivity levels in the coastal marine environment of the West African region, including Ghana. In Ghana, the few radioactivity studies in the coastal environment have focussed on beach sediments (Amekudzie et al., 2011; Nyarko et al., 2011), estuarine sediments (Mahu et al., 2016) and produce water from offshore oil fields (Kpeglo et al., 2016). Due to rapid growth of urbanisation and industrialisation in Tema, increasing maritime traffic and infrastructural expansion works at the Tema Port is likely to intensify chemical contamination in the harbour. Monitoring of such developing coastal areas is, therefore, essential to ensure that these socio-econonic and develomental activities do not adversely affect human health and the proper functioning of coastal aquatic ecosystems. The objective of this study was to assess the radioactivity concentrations of natural (^{238}U, ^{210}Pb, ^{226}Ra, ^{232}Th, ^{228}Ra, ^{228}Th and ^{40}K) and anthropogenic (^{137}Cs) radionuclides in surface sediments from the Tema Harbour in Ghana and their radiological and radioecological significance.

2. Materials and methods

2.1. Description of the study area

The Tema Harbour, situated in the Gulf of Guinea along the Ghana coast at Tema (Fig. 1), is semi-enclosed with a water area of 1.7 km^2 within a wider harbour zone of 3.9 km^2. It consists of the Main Harbour, the Fishing Harbour comprising the Inner Fishing Harbour, the Outer Fishing Harbour, and the Canoe Basin (Nyarko, Fletcher, Addo, Foli, & Mahu, 2014). Water depths range from 7.5–11.4 m for the Main Harbour, 7.5–8.5 m for the Fishing Harbour and 3.5–7.0 m for the Canoe Basin (http://www.ghanaports.gov.gh). The Main Harbour has 14 berths, a total of 4580 m breakwater, a shipyard and dry dock for ship repairs, and a 240 m wide access channel. The harbour does not receive riverine inflows, but does receive wastewater from the Tema Township. The Fishing Harbour serves as a landing site for fishing vessels, where repairs and re-fuelling of marine crafts are also carried out. A 2010 feasibility study report by Halcrow Engineers (unpublished) on the Tema Harbour revealed that it is underlain mainly by gneiss rocks composed of feldspar, quartz and micaceous minerals. The Tema Harbour is located in an industrial environment and thus, it could be impacted by industrial activities.

2.2. Sediment sampling

Grab surface sediment samples were collected in November 2013 from 21 stations within the Tema Harbour (Fig. 1), excluding rocky areas and berths where ships had docked (e.g. the area between S7 and S14). Fourteen (14) of the sampling stations were located in the Main Harbour (S1-S14), three (3) stations in the Canoe Basin (S15-S17), and two (2) stations each in the Inner Fishing Harbour (S18 and S19) and the Outer Fishing Harbour (S20 and S21). Geographical coordinates of the sampling stations were recorded using a Garmin Global Positioning System (GPS). At each station, the redox potential (E_h) was measured *in situ* near the sediment-water interface using a Hanna multi-parameter probe (HI 9829, Hanna Instruments, USA).

Sediment samples were collected using a 3.5 L Ekman bottom grab sampler. The sampling locations were approximately 200 m apart, covering a wide area and range of water depths to provide representative data on the radionuclide distribution in the harbour. To minimise potential loss of fine particles via leakage of water from the grab, it was ensured that only grabs that arrived firmly closed on the deck were sampled for analysis. In addition, only grabs that were not filled with sediment to the lid were used to assure minimal disturbance of the surface sediments.

About 100 g wet weight portions of surface sediments were taken with a clean plastic spoon into polyamide Rilsan® bags and securely closed. With extremely low potential for diffusion of materials across their surfaces (http://tub-ex.com/products/rilsan/), Rilsan® bags are suitable for collection and storage of sediment samples and they have been used in environmental monitoring programmes (Serigstad et al., 2010). To minimise contamination from the grab, sediments in direct contact with the grab were not used and the sampling spoon was washed with deionised water after each sampling. All sediment samples were kept on ice and transported to the Chemistry Laboratory at the Ghana Atomic Energy Commission (Greater Accra, Ghana) for further analyses.

2.3. Sample analyses

For the analyses of radionuclides, wet sediment samples were oven-dried at 50 °C till constant weight. Radiometric analyses of sediment samples were conducted at the ENEA S. Teresa laboratory (Italian National Agency for New Technologies, Energy and Sustainable Economic Development, La Spezia, Italy). The sediment samples were ground and placed in 5 g plastic vials of standard geometry, closed and sealed air-tight, and then stored for at least 22 days to ensure secular equilibrium between the parent nuclides and their short-lived daughter nuclides. The sealed samples were then counted for 2–3 days, and activities measured for ^{210}Pb at 46.5 keV, ^{214}Pb at 352 keV, ^{212}Pb at 239 keV, ^{208}Tl at 583 KeV, ^{228}Ac at 338 and 911.0 keV, ^{234}Th at 63.3 and 92.5 keV, ^{40}K at 1460 keV and ^{137}Cs at 662 keV. The measured activities were decay-corrected with respect to the date of sediment sampling (Sirelkhatim et al., 2008) and associated errors were determined from l-sigma counting statistics (Nyarko et al., 2011). The activity of ^{228}Th was obtained from the activity of its daughter ^{212}Pb radionuclide and ^{226}Ra and ^{228}Ra from the activities of ^{214}Pb and ^{228}Ac, respectively.

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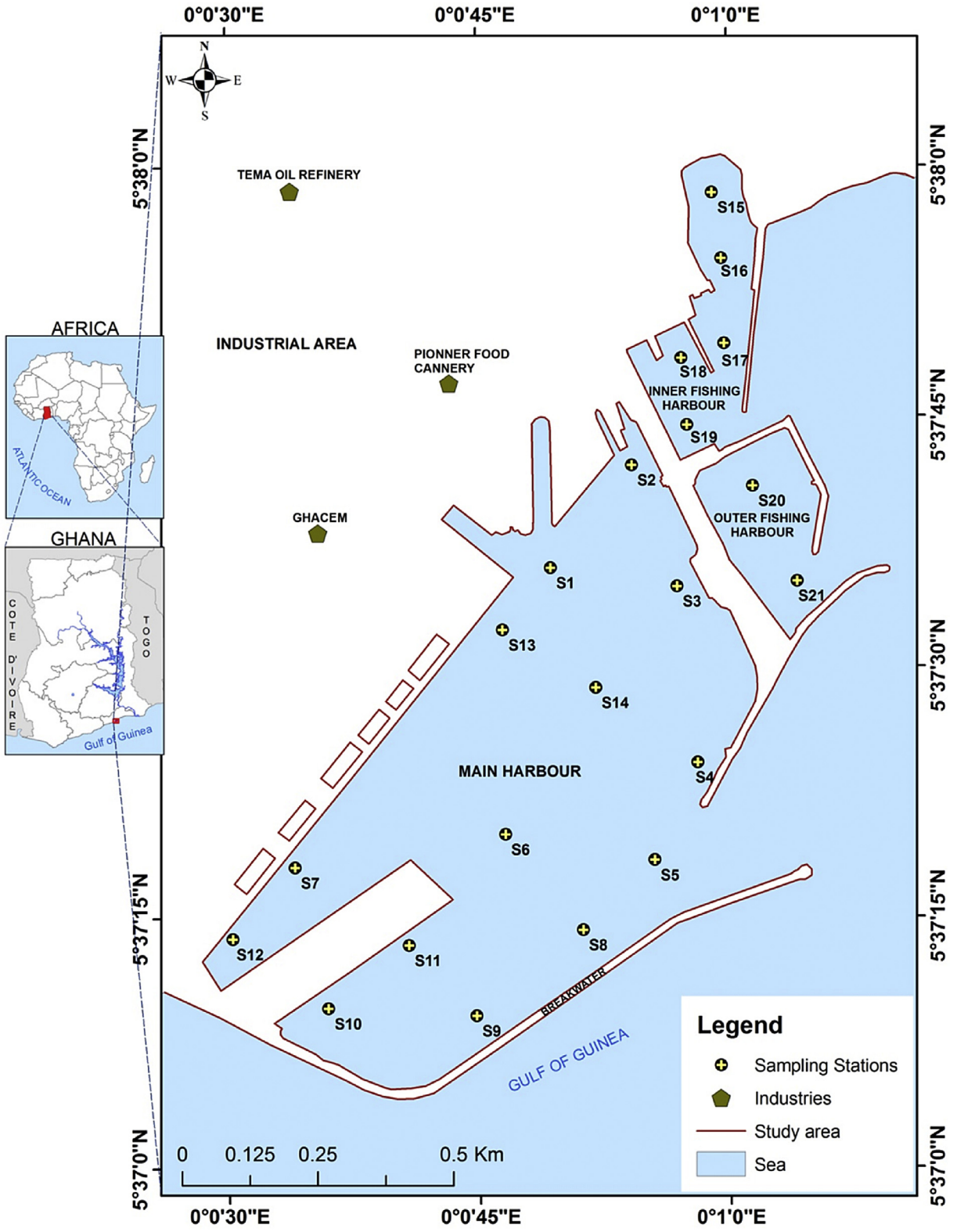

Fig. 1. Map of Tema Harbour (Greater Accra, Ghana) showing sediment sampling locations.

The average activity of ^{228}Ac and ^{212}Pb was used as a proxy for ^{232}Th activity. Since no unsupported ^{234}Th ($^{234}Th_{ex}$) was observed, the supported ^{234}Th activity was used as a proxy for ^{238}U activity.

Samples were analysed using a Gamma spectrometer coupled to an ORTEC low background intrinsic germanium coaxial detector (17.6% absolute efficiency, 1.8 keV nominal resolution at 662 keV ^{137}Cs gamma emission). Prior to the sample radioactivity analyses, the Gamma ray detectors were calibrated for measurement of ^{40}K using the IAEA-385 Certified Reference Material (CRM), while calibrations for ^{238}U series radionuclides were performed using the CANMET (Canada Centre for Mineral and Energy Technology) Reference Standard (DL1a), being a U-Th ore in which ^{210}Pb and

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^{226}Ra exist in secular equilibrium. Calibrations for ^{232}Th series radionuclides were also performed using the IAEA Reference Standard (RGTh-1) prepared by CANMET, while calibration for ^{137}Cs was performed using the Eckert & Ziegler Analytics Reference Standard (QCYA48). Quality of all results was routinely checked by analysing IAEA-300 and IAEA-315 Reference Materials as well as detector blanks (empty sample containers) processed in a similar way as the actual samples. Correction of measured activities for self-adsorption effects was done based on measurements of the attenuation of a known ^{210}Pb Gamma source by the samples.

2.4. Radiological risk assessment

Dredged radioactive-contaminated harbour sediments may be disposed of on land or used for other purposes such as building, which can potentially result in human exposure to ionising radiations and cause radiological effects. For human protection and appropriate handling of radioactive-contaminated sediments, it is essential to characterise the associated potential radiological risks. Five radiological hazard indices were estimated following established formulae to characterise the potential radiation dose to humans resulting from exposure to sediment radioactivity, *viz.* (1) total absorbed dose rate in air (D), (2) radium equivalent activity (Ra_{eq}), (3) external hazard index (H_{ex}), (4) annual gonadal dose equivalent (AGDE), and (5) annual effective dose equivalent (AEDE).

The D expresses the rate of exposure to gamma radiation in air at 1 m above the ground due to the activities of ^{226}Ra, ^{232}Th and ^{40}K in the sediment samples. Ra_{eq} is a weighted sum of the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in a sediment sample, which allows comparison with their individual ^{226}Ra, ^{232}Th and ^{40}K activity concentrations (Sugandhi et al., 2014). H_{ex} is a measure of the indoor radiation dose rate associated with external gamma radiation exposure from natural radionuclides in building materials and it is important when considering the suitability of sediments as building materials (Xinwei, Lingqing, & Xiaodan, 2006). For human health safety, the value of H_{ex} must not exceed 1.0 (Kurnaz et al., 2007; Xinwei et al., 2006). Owing to their relatively higher sensitivity to ionising radiation compared to other organs of the body, the gonads are considered to be at a high risk of radiation exposure and are therefore of great interest in radiological assessment (Kurnaz et al., 2007). The AGDE estimates the potential radiation dose that the gonads may receive from ^{226}Ra, ^{232}Th and ^{40}K. It is also usual to convert D to AEDE to assess the dose rate to an individual from outdoor gamma radiation over a period of one year.

The D and Ra_{eq} were calculated using Eqs. (1) and (2), respectively, following (El Mamoney & Khater, 2004):

$$D\ (\mathrm{nGy.h^{-1}}) = 0.462A_{Ra} + 0.604A_{Th} + 0.0417A_{K} \quad (1)$$

$$Ra_{eq}\ (\mathrm{Bq\ kg^{-1}}) = A_{Ra} + 1.43A_{Th} + 0.077A_{K} \quad (2)$$

H_{ex} was calculated using Eq. (3) following (Xinwei et al., 2006):

$$H_{ex} = (A_{Ra}/370) + (A_{Th}/259) + (A_{K}/4810) \quad (3)$$

AGDE and AEDE were calculated using Eqs. (4) and (5), respectively, following (Kurnaz et al., 2007).

$$\mathrm{AGDE}\ (\mu\mathrm{Sv.y^{-1}}) = 3.09A_{Ra} + 4.18A_{Th} + 0.314A_{K} \quad (4)$$

$$\mathrm{AEDE}\ (\mu\mathrm{Sv.y^{-1}}) = D\ (\mathrm{nGy.h^{-1}}) \times 8760\ \mathrm{h} \times 0.2 \times 0.7\ \mathrm{Sv.Gy.y^{-1}} \times 10^{-3} \quad (5)$$

2.5. Radioecological risk assessment

Radioactivity contamination in sediments may put aquatic organisms at risk of ionising radiation effects. Therefore, the ERICA tool (version 1.2) was applied to assess the potential dose rates to organisms in the harbour. A detailed description of the ERICA tool can be found in literature (Beresford et al., 2007; Brown et al., 2008; Larsson, 2008). The tool is based on data gathered from extensive radioecological and dosimetric studies and uses generalised ecosystem representations, also referred to as reference organisms (Beresford et al., 2007). In this context, a reference organism is defined as *"a series of entities that provide a basis for the estimation of radiation dose rate to a range of organisms which are typical, or representative, of a contaminated environment"* (Beresford et al., 2007).

In this study, the ERICA tool was used to estimate activity concentrations in ten (10) reference organisms captured in the ERICA database, *viz. phytoplankton*, *zooplankton*, *macroalgae*, *pelagic/benthic fishes*, *mammals*, *crustaceans*, *mollusc-bivalves*, *sea anemones*, and *polychaete worms*. In estimating the activity concentrations in the selected reference organisms, the highest measured activity concentrations in the sediments (Bq kg^{-1} dry wt.) were used as input data to represent "worst case scenarios". Default concentration ratios for the reference organisms in ERICA were then applied. Since, by default, ^{40}K was not included in the ERICA database, this isotope was also not considered here.

Furthermore, a Tier 2 ERICA assessment was conducted to estimate the total dose rates to biota, applying a default uncertainty factor of 3.0 in the ERICA tool to ensure there will be less than 5% probability of modelled dose rates exceeding the screening dose rate. Since the magnitude of biological effects varies with different types of ionising radiation (Schmid & Schrader, 2007), default ERICA weighting factors of 10.0 for alpha, 1.0 for beta/gamma, and 3.0 for low beta radiation were applied to give appropriate weights to the dose rates. As dose rate to biota is a function of the duration of exposure, a default ERICA occupancy factor of 1.0 (i.e., fraction of time that the organism spends at a specified location in its habitat) was assigned to each reference organism, assuming they spend 100% of the time at their specified locations (i.e. in the water column, on the sediment surface or inside the sediment).

3. Results and discussion

3.1. Concentrations of radionuclides in Tema Harbour sediments

The activity concentrations of ^{238}U, ^{210}Pb, ^{226}Ra, ^{228}Ra, ^{228}Th, ^{232}Th, ^{40}K and ^{137}Cs in the surface sediment samples are shown in Table 1. The activity concentrations of ^{40}K were relatively higher than those of the other radionuclides, ranging from 250 to 570 Bq kg^{-1} with a mean of 320 Bq kg^{-1}. Doyi, Oppon, Glover, Gbeddy, and Kokroko (2013) reported higher levels of ^{40}K and ^{232}Th for rocks and ore from mines in the Upper East Region of Ghana in the ranges of 950–2800 and 81–880 Bq kg^{-1}, respectively, with a higher mean ^{238}U level of 66 (± 8) Bq kg^{-1}. The levels of ^{210}Pb (except at station S15) were markedly higher than those of ^{238}U, ^{226}Ra, ^{232}Th, ^{228}Ra and ^{228}Th. Their mean value, 210 (±10) Bq kg^{-1} (~200 Bq kg^{-1} for unsupported ^{210}Pb) is to some extent higher, but comparable to the values of unsupported ^{210}Pb found by Mahu et al. (2016) in the upper layers of sediment cores sampled in the Amisa (103 Bq kg^{-1}), Sakumo II (157 Bq kg^{-1}) and Volta (123 Bq kg^{-1}) estuaries in Ghana, respectively.

In coastal marine sediments, an excess of ^{228}Th above that supported by the parents ^{232}Th or ^{228}Ra has often been reported, and used for radiometric dating (Koide, Bruland, & Goldberg, 1973). In the Tema Harbour sediments, the radioactivity concentrations of

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Table 1
Radioactivity concentrations (Bq kg^{-1} dw) in Tema Harbour surface sediments and water depth and E_h at the sampling stations.

Sampling station	Water depth (m)	E_h (mV)	^{238}U series radionuclides			^{232}Th series radionuclides			^{40}K	^{137}Cs
			^{238}U	^{210}Pb	^{226}Ra	^{232}Th[a]	^{228}Ra	^{228}Th		
S1	10	−96	30 ± 6	230 ± 10	12 ± 2	21 ± 3	18 ± 2	24 ± 1	250 ± 10	1.6 ± 0.4
S2	7	−128	39 ± 5	200 ± 10	11 ± 1	25 ± 3	24 ± 2	26 ± 1	260 ± 10	1.7 ± 0.5
S3	8.5	−100	43 ± 5	250 ± 10	13 ± 1	26 ± 4	26 ± 3	25 ± 1	270 ± 10	2.0 ± 0.5
S4	8	−70	40 ± 4	260 ± 10	18 ± 2	32 ± 4	30 ± 3	33 ± 1	350 ± 10	1.1 ± 0.5
S5	8	−65	28 ± 5	250 ± 10	14 ± 2	21 ± 3	19 ± 2	22 ± 1	250 ± 10	1.1 ± 0.5
S6	8	−94	28 ± 4	250 ± 10	17 ± 2	70 ± 4	67 ± 3	72 ± 1	330 ± 10	1.3 ± 0.5
S7	8	−90	28 ± 4	130 ± 10	13 ± 1	37 ± 3	34 ± 2	40 ± 1	460 ± 20	0.9 ± 0.5
S8	9	−80	28 ± 4	210 ± 10	13 ± 1	19 ± 3	18 ± 2	20 ± 1	270 ± 10	1.2 ± 0.4
S9	8.5	−80	35 ± 4	290 ± 10	16 ± 2	27 ± 3	24 ± 2	29 ± 1	290 ± 10	2.3 ± 0.5
S10	9.5	−78	32 ± 4	230 ± 10	15 ± 1	26 ± 3	24 ± 2	28 ± 1	350 ± 10	1.1 ± 0.4
S11	8.5	−70	21 ± 4	110 ± 10	10 ± 1	38 ± 3	36 ± 2	39 ± 1	570 ± 20	0.5 ± 0.3
S12	9	−50	47 ± 7	130 ± 10	14 ± 1	44 ± 4	44 ± 3	43 ± 1	390 ± 20	2.2 ± 0.5
S13	7.5	−70	36 ± 3	240 ± 10	17 ± 1	29 ± 3	27 ± 2	31 ± 1	330 ± 10	1.1 ± 0.4
S14	10	−95	42 ± 4	280 ± 10	16 ± 1	28 ± 3	25 ± 2	31 ± 1	330 ± 10	1.4 ± 0.4
S15	3	−400	24 ± 4	20 ± 5	16 ± 1	37 ± 3	36 ± 2	38 ± 1	330 ± 10	<0.3
S16	4	−210	26 ± 5	230 ± 10	13 ± 2	34 ± 3	35 ± 2	32 ± 1	310 ± 10	1.9 ± 0.5
S17	5	−140	32 ± 4	230 ± 10	13 ± 2	25 ± 3	21 ± 2	28 ± 1	310 ± 10	1.5 ± 0.4
S18	8	−115	42 ± 5	310 ± 20	14 ± 1	23 ± 3	22 ± 2	23 ± 1	270 ± 10	1.9 ± 0.4
S19	8	−110	35 ± 6	240 ± 10	11 ± 2	24 ± 4	23 ± 3	25 ± 1	300 ± 10	1.6 ± 0.5
S20	8	−100	27 ± 4	170 ± 10	20 ± 1	26 ± 3	24 ± 2	27 ± 1	300 ± 10	1.4 ± 0.4
S21	8	−90	41 ± 5	170 ± 10	18 ± 1	28 ± 3	27 ± 2	29 ± 1	300 ± 10	1.5 ± 0.5
Average	–	–	34 ± 5	210 ± 10	14 ± 1	30 ± 3	29 ± 2	31 ± 1	320 ± 10	1.5 ± 0.5

Errors are 1 standard deviation from counting statistics.
[a] ^{232}Th activities are estimated from grand-daughters activities.

^{232}Th, ^{228}Ra and ^{228}Th were comparable within the involved uncertainties. Activity concentrations of ^{137}Cs in the sediments were markedly low, varying from <0.3 to 2.3 Bq kg^{-1} with a mean of 1.5 Bq kg^{-1}. These values are comparable to those reported by Mahu et al. (2016) for the surface layers of estuarine sediments from Ghana (in the range 0–7 Bq kg^{-1}). The low ^{137}Cs levels suggest low atmospheric fallout in the study area, coupled with natural decay following its deposition (Livingston & Povinec, 2000; Pfitzner, Brunskill, & Zagorskis, 2004) or remobilisation from sediment into seawater (Sugandhi et al., 2014). Thus, Junge et al. (2010) have reported ^{137}Cs concentrations in the range 0.5–6.5 Bq kg^{-1} in farmland soils from Nigeria.

The levels of ^{210}Pb (20 ± 5 Bq kg^{-1}) and ^{137}Cs (<0.3 Bq kg^{-1}) in the shallow station of the Canoe Basin (S15) were very low compared to the levels found at the remaining stations. Several mechanisms are involved in the distribution of radionuclides in the harbour such as hydrodynamics, waves, tides, and vessel movements and dredging. The Canoe Basin is receives sediments from mixed sources including sand bars bordering one of its sides. In May 2013, maintenance dredging was carried out in the Canoe Basin to ensure safer navigation and increase berthing capacity for the operation of canoes. Dredging can potentially remove contaminated sediments while influx of sand may cause dilution, and may partly account for the low radioactivity levels of ^{210}Pb and ^{137}Cs at S16. In the Main Harbour, the ^{210}Pb levels at stations S7 (130 ± 10 Bq kg^{-1}), S11 (110 ± 10 Bq kg^{-1}) and S12 (130 ± 10 Bq kg^{-1}) were relatively lower than those at the other stations. This may partly be due to a dilution effect of the discharges of industrial wastewater and sand channelled into the harbour at S7, which can also affect neighbouring areas such as stations S12 and S11. On the contrary, Table 1 shows that these same three stations (S7, S11 and S12) recorded relatively higher levels of ^{40}K, possibly due to organic enrichment in sediments as a result of the waste water discharges.

The ratios of ^{238}U/^{226}Ra, ^{238}U/^{210}Pb and ^{210}Pb/^{226}Ra were in the ranges of 1.4–3.5, 0.1–0.4 and 1.3–22.4, respectively. The departures of these ratios from 1.0 indicate disequilibria, a general characteristic of marine surface sediments previously observed by others (Chen & Huh, 1999; Koide et al., 1973), which may be caused by anthropogenic influences (Al-Trabulsy et al., 2011; El Mamoney & Khater, 2004; Nyarko et al., 2011; UNSCEAR, 2000). The origin of the excess ^{210}Pb found in sediments is well known, and it is the basis of the ^{210}Pb-based radiometric method for dating recent sediments (Mabit et al., 2014). On the contrary, the observed disequilibria between ^{238}U (derived from ^{234}Th activity, see Section 2.3) and ^{226}Ra may be ascribed to differences in geochemical behaviour of the radionuclides (Koide et al., 1973). ^{226}Ra and ^{238}U are known for their different sorption and mobility characteristics; ^{226}Ra and ^{238}U are less particle-reactive than Th isotopes with greater tendency to diffuse from sediments into the surrounding seawater (Chen & Huh, 1999; Sirelkhatim et al., 2008; Sugandhi et al., 2014). The higher levels of ^{238}U relative to ^{226}Ra in the sediments may be due to diffusion and loss of Ra from sediment owing to its higher solubility in seawater or higher leachability of ^{226}Ra from the harbour sediments than ^{238}U-^{234}Th. At the time of sampling, the bottom water was anoxic, having E_h values in the range of −50 to −400 mV (Table 1), which may enhance immobilisation and precipitation of ^{238}U in the sediments (El Mamoney & Khater, 2004). Mohamed, Mahmood, Ahmad, and Ishak (2010) reported ^{228}Ra/^{226}Ra activity ratios in the range 1.2–2.9 with ^{238}U/^{226}Ra activity ratios in the range 1.1–5.6 for southern South China Sea surface sediments, which compare well with the ranges 1.4–3.9 and 1.6–3.5 found in this work (Table 1) for ^{228}Ra/^{226}Ra and ^{238}U/^{226}Ra activity ratios, respectively.

The mean radioactivity levels in the Tema Harbour sediments and levels reported for other areas are shown in Table 2. With the exception of Labadi that recorded higher levels of ^{226}Ra and ^{232}Th, the mean radioactivity levels in the Tema Harbour sediments were generally higher than those reported in sediments from other areas, i.e. Chorkor, James Town, Nungua, Kokrobite, Teshie and Weija of the Greater Accra coast of Ghana (Amekudzie et al., 2011). Nyarko et al. (2011) also reported lower activity concentrations of ^{210}Pb in beach sand samples along the Ghana coast ranging from 1.6 up to 4.5 Bq kg^{-1} and ^{137}Cs concentrations below the detection limit of 0.4 Bq kg^{-1}. The mean radioactivity levels in the Tema Harbour sediments were also higher than the levels reported for the Mediterranean coast of Egypt (Higgy, 2000), but were comparable to

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Table 2
Radioactivity levels in Tema Harbour surface sediments and levels reported in sediments from other areas of the world.

Sampling station	^{238}U series radionuclides (Bq kg^{-1} dw)			^{232}Th series radionuclides (Bq kg^{-1} dw)			^{40}K (Bq kg^{-1} dw)	^{137}Cs (Bq kg^{-1} dw)	Reference
	^{238}U	^{210}Pb	^{226}Ra	^{232}Th	^{228}Ra	^{228}Th			
Ghana coast									
Chorkor	–	–	1.42	1.49	–	–	21.31	–	Amekudzie et al. (2011)
James Town	–	–	0.82	1.04	–	–	14.67	–	Amekudzie et al. (2011)
Labadi	–	–	140.8	732.6	–	–	43.97	–	Amekudzie et al. (2011)
Nungua	–	–	4.05	8.64	–	–	41.17	–	Amekudzie et al. (2011)
Kokrobite	–	–	3.74	6.63	–	–	17.76	–	Amekudzie et al. (2011)
Teshie	–	–	2.85	9.66	–	–	61.01	–	Amekudzie et al. (2011)
Weija	–	–	0.62	0.17	–	–	8.6	–	Amekudzie et al. (2011)
Tema Habour	34	210	14	30	29	31	325	1.5	This study
Other areas									
Port Sudan, Sudan	–	–	11.05	–	10.35	–	311	7.02	Sam et al. (1998)
Sawakin Harbour, Sudan	–	–	12.61	–	6.18	–	192	4.51	Sam et al. (1998)
Patras Harbour, Greece	–	–	22.6	24.5	–	–	497	3.1	Papaefthymiou et al. (2007)
Mumbai Harbour, India	–	–	10.6		12.7		436	21.6	Sugandhi et al. (2014)
Karachi Harbour, Pakistan			23.9	–	23.5	–	527	<1.3	Akram et al. (2006)
Saudi coast, Gulf of Aqaba	16.97	–	11	22	19	–	641	3.5	Al-Trabulsy et al. (2011)
Red Sea coast, Egypt	25.5	26	24.6	–	–	–	427.5	–	El Mamoney and Khater (2004)
Mediterranean coast, Egypt	8.8	–	5	2.1	–	–	46		Higgy (2000)
Caspian Sea coast, Iran	177	–	–	117	–	–	1085	131	Abdi et al. (2009)
Malaysia coast, South China Sea	45.9		27.7	73.3	66.2	–	–	–	Mohamed et al. (2010)
World average	35[a]	25[b]	25[b,c,d,e]	25[c,e]	–	–	373[d,e]	–	[a]SureshGandhi et al. (2014); [b]Nyarko et al. (2011); [c]Qureshi et al. (2014); [d]Kurnaz et al. (2007); [e]Sugandhi et al. (2014)

levels reported for the Saudi coast of the Gulf of Aqaba, except for their higher ^{40}K (Al-Trabulsy et al., 2011); the Red Sea coast of Egypt (El Mamoney & Khater, 2004), Port Sudan and Sawakin Harbour of Sudan (Sam et al., 1998), Patras Harbour, Greece (Papaefthymiou et al., 2007), Mumbai Harbour, India (Sugandhi et al., 2014), and the Karachi Harbour, Pakistan (Sugandhi et al., 2014). The mean radioactivity levels of ^{238}U, ^{232}Th, ^{40}K and ^{137}Cs in the Tema Harbour sediments were, however, much lower than those found in polluted sediments from the Caspian Sea coast of Iran (Abdi, Hassanzadeh, Kamali, & Raji, 2009). Mohamed et al. (2010) found comparatively higher levels of ^{226}Ra, ^{228}Ra, ^{238}U and ^{232}Th for surface sediments from the Malaysia coast of the southern South China Sea. Generally, the mean radioactivity levels in the Tema Harbour sediments were comparable to the reported worldwide averages (Nyarko et al., 2011; Papaefthymiou et al., 2007; Qureshi et al., 2014; SureshGandhi et al., 2014), except for ^{210}Pb which was almost an order of magnitude higher than its grandparent ^{226}Ra, due to the presence of unsupported ^{210}Pb.

3.2. Radiological significance of Tema Harbour sediment radioactivity levels

The calculated hazard indices for the Tema Harbour sediments and hazard indices reported for sediments from other areas of the world are presented in Table 3. The ranges (means) of D, Ra_{eq}, H_{ex}, AGDE, and AEDE for the Tema Harbour sediments were 29–64 nGy.h^{-1} (39 nGy.h^{-1}), 61–142 Bq kg^{-1} (82.7 Bq kg^{-1}), 0.2–0.4 (0.2), 203–447 μSv.y^{-1} (273 μSv.y^{-1}), and 35–78 μSv.y^{-1} (47 μSv.y^{-1}), respectively. These mean D, Ra_{eq}, H_{ex}, AGDE, and AEDE values were below the recommended values, although there were a few areas where the measured Ra_{eq} (i.e. S4, S6, S7, S11, S12 and S15), ADGE (i.e. S6, S7, S11, S12 and S15) and AEDE (i.e. S6) exceeded the recommended values. Thus, the radioactivity levels in the harbour sediments are generally of little radiological concern for human health.

Compared to this study, Amekudzie et al. (2011) reported higher mean D (77 nGy.h^{-1}) and H_{ex} (0.5), but lower mean Ra_{eq} (9 Bq kg^{-1}) values for sediments from Chorkor, James Town, Nungua, Kokrobite, Teshie and Weija along the Greater Accra coast of Ghana (Table 3). Comparable mean values of D (42 nGy.h^{-1}) and Ra_{eq} (101 Bq kg^{-1}) were reported for the Red Sea coast of Egypt by El Mamoney and Khater (2004), while a lower mean D value of 5.5 nGy.h^{-1} was reported for the Mediterranean coast of Egypt by Higgy (2000). Higher mean values of D (63 nGy.h^{-1}), Ra_{eq} (176 Bq kg^{-1}) and H_{ex} (0.5) were reported for sediments from the Caspian Sea coast by Abdi et al. (2009), while a lower mean H_{ex} value of 0.1 was reported for the Saudi coast of the Gulf of Aqaba by Al-Trabulsy et al. (2011).

3.3. Radioecological significance of Tema Harbour sediment radioactivity

Table 4 presents the estimated radioactivity levels in the selected reference organisms. Generally, biota exhibited potentially higher accumulation levels of ^{210}Pb (1.18–570 Bq kg^{-1}) relative to the other radionuclides (0–0.05 for ^{137}Cs; 0.82–14.4 for Ra; 0.01–6.60 for Th, and 0.06–17.5 Bq kg^{-1}for ^{238}U), possibly reflecting the relatively higher levels of ^{210}Pb in the sediments. Thus, sediments may be an important source of ^{210}Pb exposure to biota. The levels of ^{210}Pb were markedly high in phytoplankton, exceeding the levels in the sediments. This is an indication of a high potential for ^{210}Pb bioaccumulation by phytoplankton as has been reported by Hassona et al. (2008). Overall, phytoplankton exhibited the highest potential for radionuclide exposure from sediment, suggesting that it could be a good bioindicator for monitoring of radionuclide contamination in the Tema Harbour. Apart from the ^{210}Pb levels in phytoplankton, the radioactivity levels in all the reference organisms were generally low compared to the levels in the sediments. This indicates that the radionuclides associate mainly with the sediments, despite their potential for bioaccumulation (Hassona et al., 2008; Sirelkhatim et al., 2008). This supports the view that sediments are the major sinks for radionuclides in aquatic ecosystems (Sugandhi et al., 2014). The very low activity levels of Th and ^{137}Cs in biota suggest that sediments may

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Table 3
Calculated D, Ra_{eq}, H_{ex}, AGDE and AEDE for Tema Harbour surface sediments and sediments from other parts of the world as well as recommended values.

	D (nGy.h^{-1})	Ra_{eq} (Bq kg^{-1})	H_{ex}	AGDE (μSv.y^{-1})	AEDE (μSv.y^{-1})	Reference
Tema Harbour						
S1	29	61	0.2	203	35	This study
S2	31	67	0.2	220	38	
S3	33	70	0.2	232	40	
S4	42	90	0.2	297	51	
S5	29	63	0.2	208	36	
S6	64	142	0.4	447	78	
S7	48	101	0.3	339	58	
S8	29	61	0.2	204	35	
S9	36	76	0.2	251	44	
S10	37	79	0.2	265	46	
S11	51	108	0.3	367	63	
S12	49	106	0.3	348	60	
S13	39	84	0.2	277	48	
S14	38	82	0.2	270	47	
S15	44	94	0.3	308	53	
S16	39	85	0.2	278	48	
S17	34	72	0.2	240	41	
S18	31	67	0.2	222	38	
S19	32	68	0.2	229	39	
S20	37	80	0.2	263	46	
S21	38	81	0.2	267	46	
Mean	39	83	0.2	273	47	
Mean values for other areas						
Caspian Sea coast, Iran	63	176	0.5	–	–	Abdi et al. (2009)
Red Sea coast, Egypt	42	101	–	–	–	El Mamoney and Khater (2004)
Saudi coast, Gulf of Aqaba	–	–	0.1	–	–	Al-Trabulsy et al. (2011)
Mediterranean coast, Egypt	5.5	–	–	–	–	Higgy (2000)
Ghana coast	77	9	0.5	–	0.1	Amekudzie et al. (2011)
Recommended value	55[a,b]	89[a]	1.0[c]	300[c]	70[c]	

D (total absorbed dose rate in air); Ra_{eq} (radium equivalent activity); H_{ex} (external hazard index); AGDE (annual gonadal dose equivalent); AEDE (annual effective dose equivalent); [a]Kurnaz et al. (2007); [b]Abdi et al. (2009); [c]Xinwei et al. (2006); -: not reported.

Table 4
Highest radioactivity concentrations in Tema Harbour sediments and ERICA-derived radioactivity concentrations in reference organisms.

	Sediment (Bq kg^{-1} dw)	Reference organisms (Bq kg^{-1} fresh weight)								
		Phytoplankton	Zooplankton	Macroalgae	Pelagic/Benthic fishes	Mammals	Crustaceans	Mollusc-bivalves	Sea anemones	Polychaete worms
^{137}Cs	2.3	0	0.03	0.02	0.02	0.05	0.01	0.01	0.05	0.04
[a]Ra	67	14.4	1.02	1.13	1.76	2.06	1.08	0.82	1.76	1.76
^{210}Pb	314	570	20	1.18	38.9	22.4	24.8	7.43	38.9	47.2
[b]Th	72	6.60	0.06	0.04	0.01	0.02	0.35	0.02	0.02	0.02
^{238}U	47	3.85	0.07	1.47	0.16	0.16	0.06	0.57	17.5	17.5

[a] Ra is for both ^{226}Ra and ^{228}Ra.
[b] Th for both ^{232}Th and ^{228}Th.

not be a major exposure pathway of these radionuclides to biota in the harbour.

The ERICA-derived dose rates to biota are presented in Table 5. It shows that phytoplankton could potentially receive higher dose rates from sediment radioactivity than the other reference organisms, which corresponds to its higher bioaccumulation potential for radionuclides (Table 4). The total dose rate to phytoplankton slightly exceeded the typical maximum value in the ERICA database (see Table 5). Whereas the total dose rate to pelagic fishes was similar to the typical minimum, that of crustaceans slightly exceeded the typical minimum value, but fell below the typical average value. In the case of zooplankton, macroalgae, benthic fishes, mammal, mollusc-bivalves and polychaete worms, the total dose rates fell below their ERICA typical minimum values (Table 5). The total dose rates to all the reference organisms fell below the screening dose rate of 400 μGy.h^{-1}, proposed by the International Atomic Energy Agency (IAEA) and the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) as the dose rate below which harmful effects are unlikely to occur in organisms.

It is worthy to note that the ERICA tool was used in this study as a screening tool to provide estimates of the radioactivity levels in biota and associated total dose rates rather than their accurate prediction. Furthermore, the estimated total dose rates to biota may be underestimated since they were based on the radioactivity concentrations of only ^{238}U, ^{210}Pb, ^{226}Ra, ^{228}Ra, ^{228}Th, ^{232}Th and ^{137}Cs. Measurement of radioactivity levels in different environmental matrices such as sediments, water and biota from the harbour will be important to validate the predictive ability of the ERICA modelling tool for the Tema Harbour ecosystem and is, therefore, recommended for future studies.

4. Conclusions

The radioactivity levels of ^{238}U, ^{210}Pb, ^{226}Ra, ^{232}Th, ^{228}Ra, ^{228}Th, ^{40}K and ^{137}Cs in surface sediment from the Tema Harbour in Ghana

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Table 5
ERICA-derived total dose rates to reference organisms and typical maximum, minimum, and average values ($\mu Gy.h^{-1}$) in the ERICA database.

Reference organism	Total Dose rate	Typical average dose rate	Typical minimum dose rate	Typical maximum dose rate
Phytoplankton	2.11	0.38	0.13	2.00
Zooplankton	0.06	0.94	0.25	5.20
Pelagic fish	0.09	0.42	0.08	3.70
Mammal	0.10	1	0.23	5.80
Macroalgae	0.13	0.87	0.52	1.40
Benthic fish	0.12	0.58	0.24	1.20
Crustacean	0.16	0.59	0.12	1.90
Mollusc-bivalve	0.09	2	0.98	5.60
Sea anemones	0.54	4.20	1.90	8.80
Polychaete worm	0.58	1.6	0.94	2.5

have been assessed for the first time. The levels of ^{40}K and ^{210}Pb were relatively higher than the levels of the other radionuclides in the Tema Harbour sediments. Large disequilibria between ^{238}U and ^{226}Ra were found, attributable to the dynamics of the radionuclides in the harbour. Apart from ^{210}Pb, the radioactivity concentrations of the natural radionuclides in the Tema Harbour sediments were comparable to worldwide average values. Evaluation of total absorbed dose rate in air (D), radium equivalent activity (Ra_{eq}), external hazard index (H_{ex}), annual gonadal dose equivalent (AGDE) and annual effective dose equivalent (AEDE) indicate that the potential dose rates to human from the sediment radioactivity levels may not present significant risks to human health. Moreover, the potential dose rates to biota derived from the ERICA assessment indicate low ecological risks associated with the radioactivity levels in the harbour sediments. This study provides baseline information on radioactivity levels in the Tema Harbour sediments for comparison to future monitoring studies.

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