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Baseline

Baseline survey of sediment contamination with ^{210}Polonium in three peri-urban creeks of Mombasa, Kenya

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ABSTRACT

^{210}Po in marine sediment from three peri-urban creeks in Mombasa was analyzed by alpha spectrometry in order to determine its baseline contamination levels. The mean ^{210}Po activity in marine sediment from Mtwapa, Tudor and Makupa creeks were 18.67 ± 1.8, 21.17 ± 2.2 and 13.09 ± 1.6 Bq Kg^{-1} dw, for surface sediment and 37.56 ± 2.14, 28.64 ± 2.86 and 30.42 ± 2.1 Bq Kg^{-1} dw respectively, for sediment cores. Comparison of ^{210}Po activity in surface sediment indicated that ^{210}Po in the creeks could be originating from the same source. However, comparison of activities in the entire cores from the three creeks indicated that ^{210}Po sources could have been different in the past. ^{210}Po activities in this study were relatively elevated compared to marine environments under similar anthropogenic influence and there is need to understand its source and fate.

Sediment is a major sink for natural and anthropogenic radionuclides and has been utilized to provide long-term pollution status of the aquatic environments (Iwegbue et al., 2007; Pandit et al., 2014). The accumulation of radionuclides in the sediment may lead to contamination of the marine environment (Hashim et al., 2004) thus the emerging need for studies to determine their spatio-temporal distribution and levels in the marine environment (Long and Chapman, 1985). Among the natural radionuclides, the alpha emitters (e.g. ^{210}Po, ^{209}Bi, ^{215}At, ^{218}Rn, ^{221}Fr, ^{223}Ra, ^{227}Th, ^{233}U, ^{238}Pu, ^{241}Am, ^{244}Cm, ^{249}Cf) have been reported to have significant radiological effects when accumulated in organisms (Skwarzec and Jahnz, 2007).

^{210}Po is a naturally occurring alpha-emitting radionuclide present in very low concentrations in the environment as a part of the uranium decay chain. It is ubiquitously distributed in rocks, soils, making up earth's crust, in the atmosphere and in natural waters (Matthews et al., 2007). ^{210}Po has a radioactive half-life of 138.4 days and is produced in the marine environment from the decay of ^{210}Pb (which is a daughter isotope derived from ^{226}Ra dissolved in seawater). ^{210}Pb can also be introduced into the marine environment directly from the atmosphere through the decay of ^{222}Rn (Skwarzec and Jahnz, 2007; Thi Van et al., 2016). It can also be derived in insignificant quantities from lead-containing wastes from uranium, vanadium, and radium refining operations (https://www.who.int/ionizing_radiation/pub_meet/polonium210/en/). The activity of ^{210}Po in the environment has increased in the past due to human activities such as fossil fuel combustion, use of phosphate fertilizers in agriculture and discharge of domestic and industrial sewage (Hariprasad and Dayananda, 2013).

^{210}Po and ^{210}Pb strongly bind to particle surfaces in the marine environment, where they can accumulate to relatively high levels in both sediment and aquatic organisms (Rožmarić et al., 2012; Skwarzec et al., 2016) thus contributing to radiation exposure to humans. For instance, ^{210}Po emits many alpha particles that may result in a relatively large biological damage (Stralberg et al., 2003). ^{210}Po is also an important radionuclide for tracing environmental processes. Even though the activity and inventory of ^{210}Pb has extensively been studied in lake and ocean sediment in order to determine sediment geochronology (Masqué et al., 2002), only a few studies have investigated ^{210}Po activities in sediment (Pempkowiak et al., 2002; Wang and Cornett, 1993).

Mombasa City (like many other coastal cities in the developing nations) is continuously experiencing an increase in urban growth, mining and industrial development thus putting pressure on marine resources and their ecosystems services particularly in the peri-urban creeks of Mtwapa, Tudor and Makupa (Okuku et al., 2011). Currently, a larger proportion of pollutants originate from land-based activities and are discharged into the creeks in the form of dissolved and particulate fluxes i.e. from domestic, industrial and hospitals effluent (Okuku et al., 2011), urban runoff, mineral sand mining, chemical weathering, combustion of fossil fuel and from dumpsites. Despite these potential sources of ^{210}Po, there is no known study that has been carried out in Kenya to determine ^{210}Po contamination. This study aimed at determining ^{210}Po activity in the sediment collected from Mtwapa, Tudor and Makupa creeks in order to understand ^{210}Po contamination (see Fig. 1).

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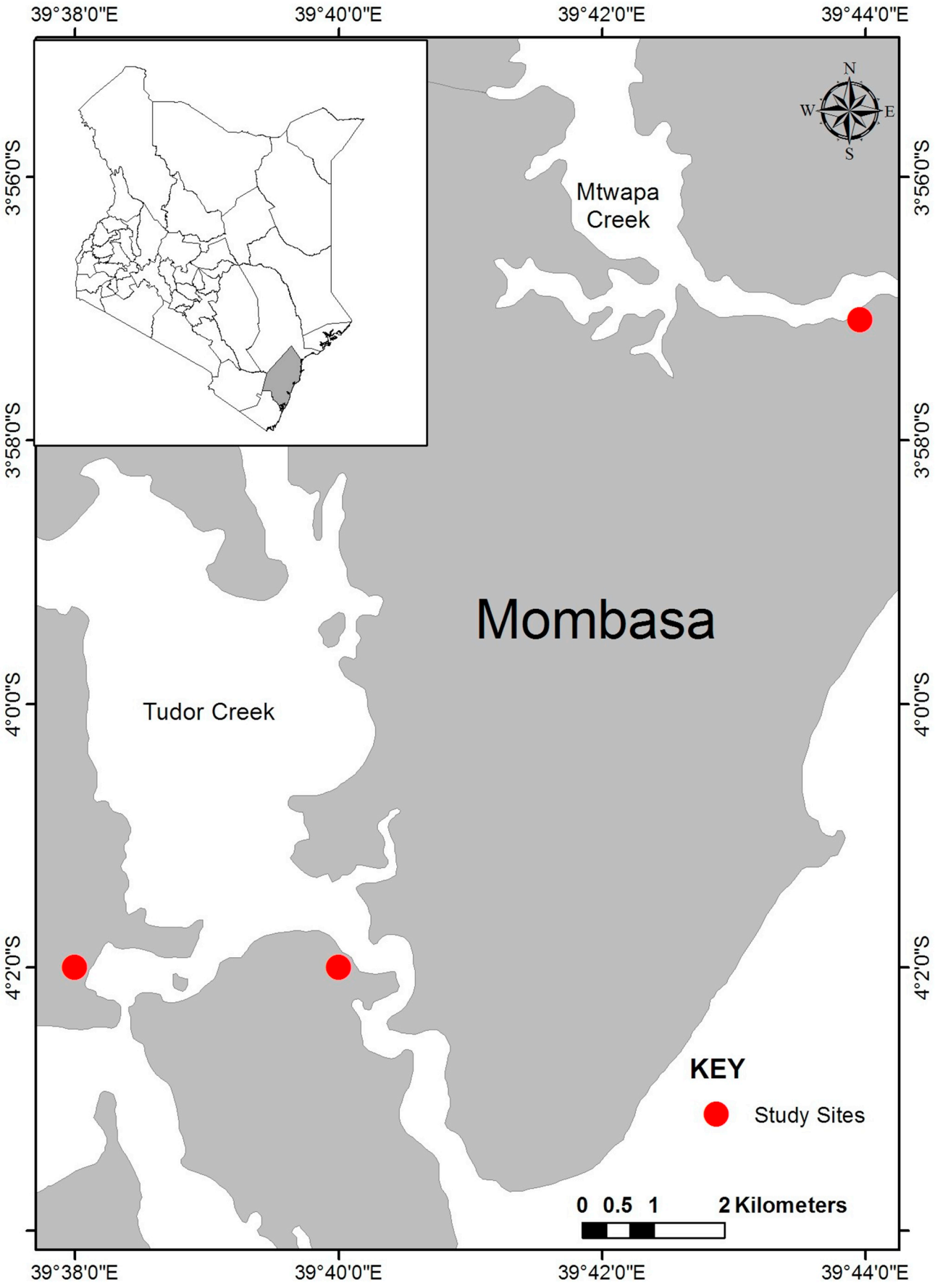

Fig. 1. Map showing the different sampling sites in the Mombasa peri-urban creeks.

Sediment cores were collected between May and June 2014 from -Tudor (4°2′ S, 39°40′ E); Makupa (39°38 E, 4°02 S, and Mtwapa (3° 55′ 00″ S) creeks using an Uwitech corer (fitted with a 100 cm plastic tube, Ø 8 cm). The cores were sectioned at a resolution of 1 cm for the top 5 cm section and thereafter at 2 cm. The sections were placed in clean polyethylene zip lock bags and stored in a cooler box to limit biological and chemical activities. The samples were then transported to Kenya Marine and Fisheries Research Institute (KMFRI) laboratory and kept frozen at −20 °C prior to analysis.

In the laboratory, the samples were weighed, oven dried at 70 °C to complete dryness and reweighed to get the moisture content. Organic matter was determined by ashing 10 g of sediment at 450 °C for 6 h. Sediment grain size was determined granulometrically using Malvern Mastersizer 2000 laser diffraction particle size analyzer. The percentage particle distribution of Mtwapa, Tudor and Makupa creeks sediment were clustered in to clay (< 3.9 μm), silt (3.9–62.5 μm) and sand (> 63 μm) according to Wentworth (1922) grain size classification.

The total ^{210}Po activity in the sediment samples was determined as described by Benmansour et al. (2006). In brief, about 0.5 g of dry, homogenized samples were weighed and put in acid-cleaned Teflon beakers. The samples were then spiked with 200 μL of ^{209}Po tracer and digested using concentrated HCl, HNO_3 and HF at 80 °C to incipient dryness. 9 g of boric acid and 100 mL of concentrated HCl was added to the residue and digested at 80 °C to incipient dryness to remove fluoride complexes. 80 mL of 0.5 M HCl was added to the residue and filtered using a GFF filter paper. ^{210}Po was plated on a copper disc at 80 °C for 6 h in a solution of HCl filtrate and ascorbic acid (added to reduce Fe^{3+} to Fe^{2+}).

^{210}Po activity was measured using alpha-spectrometry with silicon surface barrier detectors (EG&G) coupled to a PC running Maestro TM data acquisition software (Benedik and Vrecek, 2001). The samples were analyzed together with reagent blacks and IAEA 327 reference materials (radionuclides in soil). The recoveries of the reference material were between 95 and 110%.

The results showed that Mtwapa creek had a higher mean (± SE) organic matter (44.54 ± 0.42%) and dry bulk density (0.939 ± 0.054 gcm^{-3}) in the surface sediments compared to Tudor (46.16 ± 0.36% and 0.800 ± 0.057 g cm^{-3}) and Makupa (12.97 ± 0.30% and 0.888 ± 0.097 g cm^{-3}) creeks, respectively. In the entire sediment core, Tudor creek had the highest organic matter (49.24 ± 0.6049.24 ± 0.60) and the least dry bulk density (0.65 ± 0.02 g cm^{-3}) as shown in Table 1. This suggests a disturbed sediment in Tudor probably due to bioturbation as discussed in studies elsewhere by Winston and Anderson (1971) and Remaili et al. (2017). Mtwapa creek had more stable and compact sediment comprising a higher clay and silt content (68.96 ± 3.38%) as reported elsewhere by Fernandes et al. (2011). The variation in sediment dry bulk density and porosity could also be a result of lower layers being compacted by the weight by upper sediment layers (Glew et al., 2001).

The mean activities of ^{210}Po in the surface sediment were highest in Tudor creek compared to Mtwapa and Makupa creeks (Table 2). The ^{210}Po activity in the surface sediment in Makupa creek was lower than in the subsequent sections suggesting deposition of recent sediment with low ^{210}Po activity from terrestrial origin mainly from ongoing land reclamation in the creek.

Table 2
^{210}Po mean (± SE) activity (Bq Kg^{-1}) in surface sediment and sediment core samples.

Creek	^{210}Po mean activity in the top 5 cm surface sediment	^{210}Po mean activity in the whole sediment core
Mtwapa	18.67 ± 1.80 Bq kg^{-1} dw	37.56 ± 2.10 Bq kg^{-1} dw
Tudor	21.17 ± 2.20 Bq kg^{-1} dw	28.64 ± 2.80 Bq kg^{-1} dw
Makupa	13.09 ± 1.60 Bq kg^{-1} dw	30.42 ± 2.10 Bq kg^{-1} dw

The high ^{210}Po activities in surface sediment (Table 2) in Tudor creek among the three creeks could be attributed to the fact that the creek is receiving relatively higher amounts of untreated wastewater from both industrial and domestic sources. Even though wastewater treatment remediates the water quality, sediment does not respond to this treatment as fast as water column (Saçan et al., 2010) resulting in sediments serving as sinks for ^{210}Po and most radionuclides/contaminants in general. Recent developments have seen various sections of the creek water masses being reclaimed by terrestrial soil that could already be contaminated with ^{210}Po radionuclide. This is in addition to sand harvesting activities in the creek, which could have also contributed to the observed activity of ^{210}Po in sediment.

A comparison of ^{210}Po activity in the surface sediment (Table 3) with other regions in the world showed that the ^{210}Po activity ranges (13.09–21.17 Bq kg^{-1}) in the three peri-urban creeks were lower than ^{210}Po activity ranges in Kapar coastal, Malaysia (13.74–68.59 Bq kg^{-1}) due to coal burning (Alam et al., 2015), Venice lagoon, Italy (26.0–45.0 Bq kg^{-1}), due to phosphogypsum stockpile (Jia et al., 2003) and in the coastal area of Syria (7.3–170.0 Bq kg^{-1}) due to loading of phosphate ore cargos into ships (Al-Masri et al., 2002). However, ^{210}Po activities reported in this study were higher as compared to other studies done elsewhere (Table 3).

When considering the entire core, the highest mean activity of ^{210}Po was observed in Mtwapa (37.56 ± 2.14 Bq kg^{-1}) followed by Makupa (30.42 ± 2.16 Bq kg^{-}1) and finally Tudor creek (28.64 ± 2.86 Bq kg^{-1}) which were significantly different ($F = 8.00$; $p < 0.01$) in the three peri-urban creeks of Mombasa city (Table 2). ^{210}Po activities along the sediment core ranging from 7.18–43.43 Bq Kg^{-1} (Tudor), 12.80–37.99 Bq Kg^{-1} (Mtwapa) and 3.82–36.55 Bq Kg^{-1} (Makupa) were lower compared to levels reported elsewhere by Faganeli et al. (2016) and Pandit et al. (2014) with ranges between 42 and 107 Bq Kg^{-1} and 15.3–88.9 Bq Kg^{-1}, respectively

The sediment cores from the three creeks showed a decrease in ^{210}Po activities with increasing depth with the highest activities observed in the surface sediment (Figs. 2, 3, and 4). The high ^{210}Po activity in the surface sediments of the vertical profile could be attributed to the unsupported fallout of ^{210}Po during wet precipitation and also due to

Table 1
Mean (± SE). Physical properties of surface sediment and whole core sediement collected from the three peri-urban creeks of Mombasa.

Sample	Parameters	Mtwapa Creek	Tudor Creek	Makupa Creek
Surface Sediment	Moisture content (%)	44.54 ± 0.42	46.16 ± 0.36	38.64 ± 0.94
	Organic matter (%)	15.74 ± 0.35	15.80 ± 0.45	12.97 ± 0.30
	Dry bulk density (g/cm^3)	0.939 ± 0.054	0.800 ± 0.057	0.888 ± 0.097
	Porosity (Φ)	0.445 ± 0.004	0.462 ± 0.004	0.386 ± 0.009
	Clay + Silt (%)	70.68 ± 4.13	50.33 ± 10.69	35.07 ± 8.64
	Sand (%)	29.32 ± 1.85	49.67 ± 4.78	64.93 ± 3.87
Sediment core	Moisture content (%)	41.69 ± 0.76	49.24 ± 0.60	23.46 ± 1.41
	Organic matter (%)	14.04 ± 0.48	15.68 ± 0.41	6.11 ± 0.54
	Dry bulk density (g/cm^3)	1.06 ± 0.04	0.65 ± 0.02	1.25 ± 0.05
	Porosity (Φ)	0.42 ± 0.01	0.49 ± 0.01	0.23 ± 0.01
	Clay + Silt (%)	68.96 ± 3.38	50.34 ± 1.29	44.75 ± 2.28
	Sand (%)	31.04 ± 3.38	49.66 ± 1.29	55.25 ± 2.28

Table 3
Comparison of ^{210}Po mean activities in surface sediment samples from different regions of the world.

Location	^{210}Po activity (Bq Kg^{-1}, dry wt)	References
Flamingo bay, Sudan	1.6–12.5	Sirelkhatim et al. (2008)
Point Calimere Coast, India	1.9	Suriyanarayanan et al. (2008)
Kudankulam Coast, India	2.3	Macklin et al. (2014)
Periyakalpattu to Paringepettai, India.	2.4–6.6	Harikrishnan et al. (2016)
Kapar coastal, Malaysia	13.7–68.6	Alam et al. (2015)
Venice lagoon, Italy	26–45	Jia et al. (2003)
Izmir Bay (Aegean Sea-Turkey)	43–132	Saçan et al. (2010)
Coastal area of the Syrian coast	7.3–170	Al-Masri et al. (2002)
Present study	13.09–21.17	

supported ^{210}Po present in the pore spaces of the sediment core formed due to the decay of radon atoms as explained by Prakash et al. (2017).

Mtwapa creek ^{210}Po activity in the upper 10 cm of the sediment core recorded fluctuating variations in concentrations in a decreasing trend after which there was a significant decrease to the 30 cm as seen in Fig. 3. Worth noting was the increase in ^{210}Po activity at 36 cm depth possibly from increased sedimentation at this depth as compared to the other sections of the sediment core. The ^{210}Po activity decreased with increasing depth in the rest of the sediment core and could be attributed to in situ decay (Matthews et al., 2007).

The high ^{210}Po activity in surface sediments of Tudor and Mtwapa creeks in comparison to Makupa creek could be due to the large clay and silt content (Table 1), as fine grain size fractions offer a larger surface area for the adsorption of this particle reactive radionuclide. Andrew et al. (2017), Chen and Torres (2018) and Gopal et al. (2018) similarly reported that ^{210}Po (among other radionuclides) is usually primarily adsorbed on clay and differential movement of fine particles could alter its profiles during the decomposition of the organic phases. Furthermore, both Tudor and Mtwapa creeks receive large amounts of untreated wastewater from industrial, agricultural and domestic sources as reported by Okuku et al. (2011), which could be contributing significantly to the sediment organic matter as reported in studies by Saçan et al. (2010) and Venunathan and Narayan (2016).

The high ^{210}Po mean activity (37.56 Bq kg^{-1}) in Mtwapa creek sediment core could be as a result of the seasonal river (River Mto Mkuu) which flows into the creek discharging finer sediment from the mainland with naturally weathered material and anthropogenic materials from agriculture activities. Similar observation was made by Aközcan and Uğur (2013) who also reported high levels of ^{210}Po in Didin due to the discharge of Buyuk Menderes River. In addition, high levels of silt and organic matter (Table 1) were found in Mtwapa creek sediment, which may have also contributed to the observed high levels of ^{210}Po activity. This is in agreement with the work by Fernandes et al. (2011) and Venunathan and Narayan (2016) who reported that sediment rich in organic matter and silt content tend to have higher ^{210}Po activities.

Mtwapa creek sediment core ^{210}Po activities showed a significant negative correlation with moisture content, dry bulk density and sand, whereas there was a significant positive correlation with porosity and silt. This is well explained by Sommerfield and Nittrouer (1999) and Abril (2011) who showed that there is a general relationship between sediment particles size and ^{210}Pb activity a progenitor of ^{210}Po (i.e., the initial activity at the tops of sediment cores) and the dependence of the hydraulic pressure has been documented, with higher activities associated with finer sediment.

Tudor creek reported the lowest ^{210}Po activities in the sediment core compared to the other two creeks (Table 2) which could be due to high sand content (≈50%) (Table 1) resulting in reduced adsorption of ^{210}Po. Tudor sediment core ^{210}Po activity showed an increase at 2 cm, a decrease to the 5th cm and a gradual increase to the 10 cm then varied activity to the 20 cm depth which could be attributed to sand harvesting activities in Tudor creek. Even though there was a decrease in ^{210}Po activity from the 20 cm down the core, ^{210}Po activity ranged between 12.801 ± 1.66 and 27.119 ± 2.37 Bq Kg^{-1} dw inferring that mixing down the sediment core which resulted from recently contaminated soils finding their way into deeper depths/layers along the sediment core (Fig. 3). The ^{210}Po activities in Tudor may be linked to sewage effluent from domestic and hospitals sources.

Makupa sediment core ^{210}Po activity decreased with increasing depth to 50 cm after which there was an increase in ^{210}Po activity (Fig. 4). This could imply disturbance of the core during sedimentation in section 47–69 cm that had a significant increase of organic matter. Chen et al. (2017) attributed such down core distribution to post-depositional nuclide migration, smearing by coring, mixing and slumping due to gas bubbling and variations in sedimentation rate.

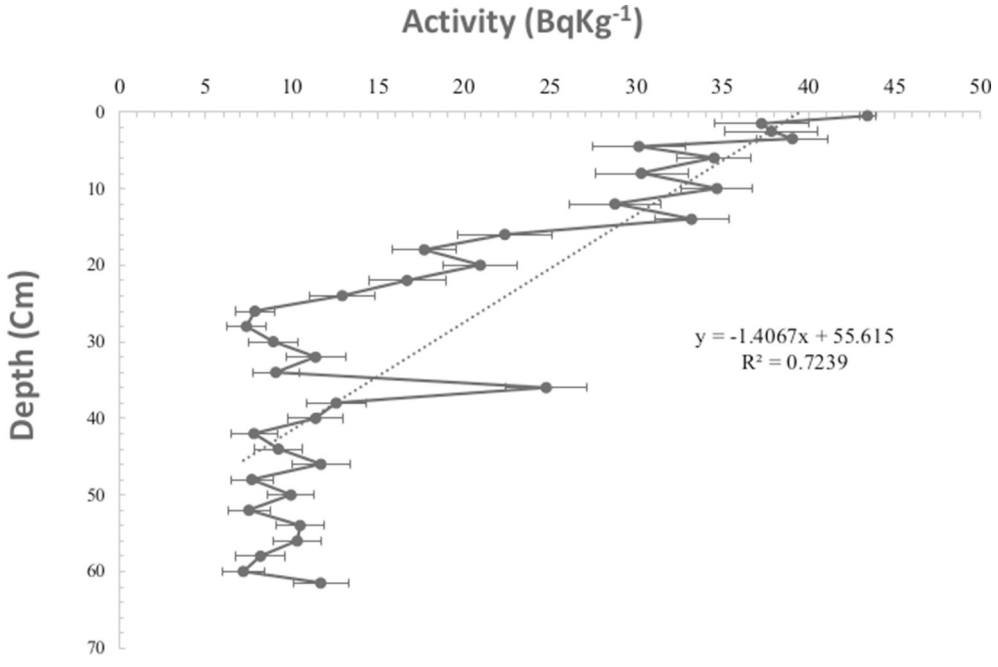

Fig. 2. Mtwapa mean (± SE) ^{210}Po activity (Bq Kg^{-1}) along the sediment core.

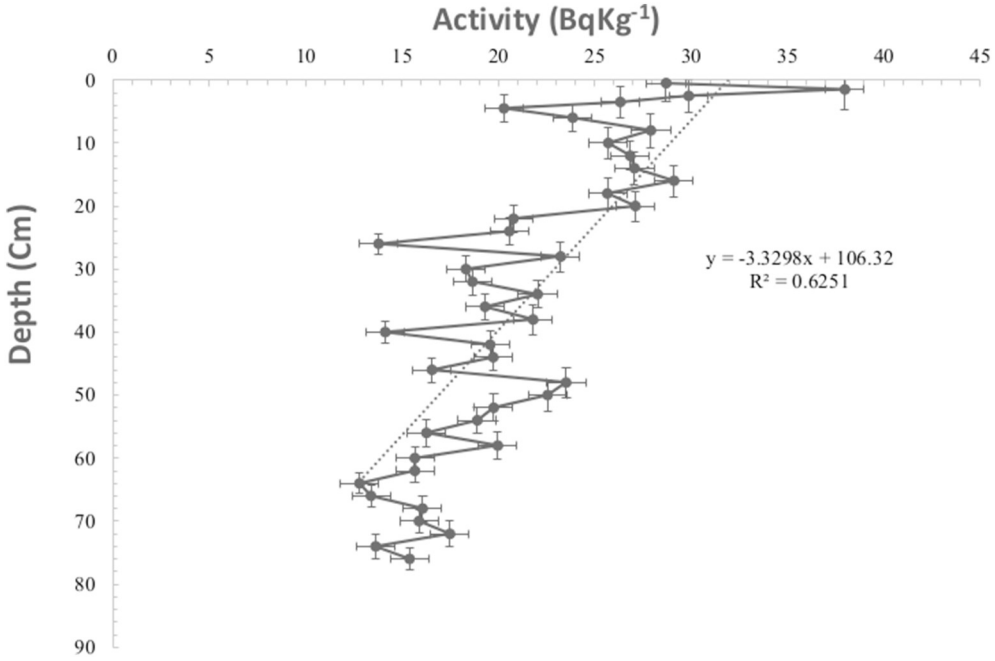

Fig. 3. Tudor sediment core mean (± SE) ^{210}Po activity (Bq Kg^{-1}).

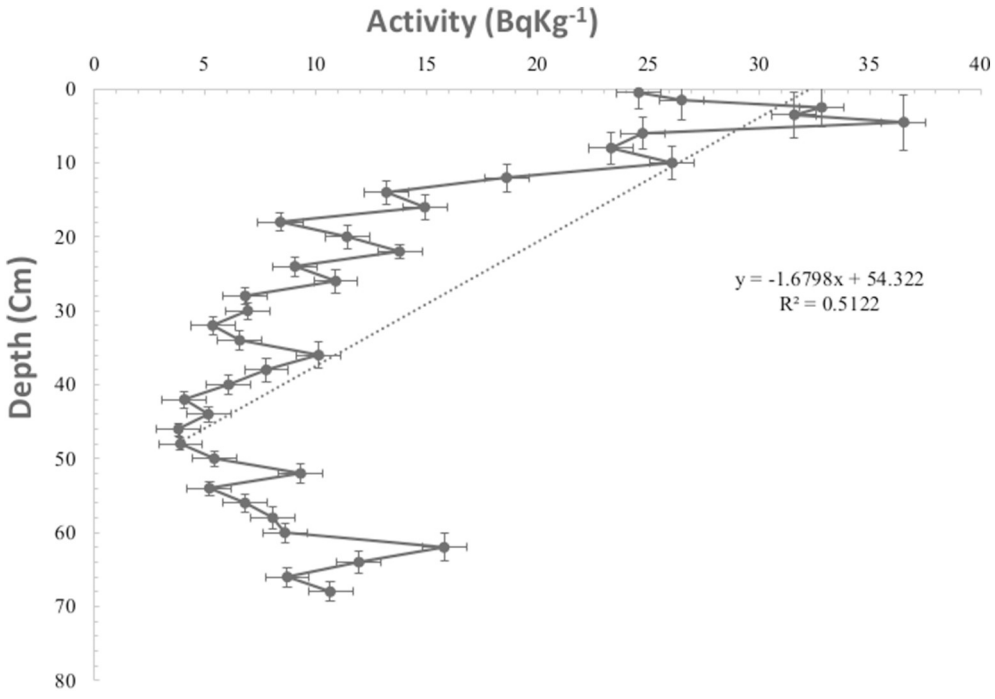

Fig. 4. Makupa mean (± SE) ^{210}Po activity (Bq Kg^{-1}) along the sediment core.

The distribution of ^{210}Po radionuclide in the sediment core in this study was probably controlled by sediment organic matter and grain size, which plays an important role as the geochemical carrier of this radionuclide. Whereas in Mtwapa creek, the ^{210}Po activities could be influenced by the seasonal river that drains into the creek in addition to wastewater inputs, for Makupa creek (even though it receives untreated wastewater), the adjacent dumpsite could be contributing to its sediment core ^{210}Po activities.

In conclusion, ^{210}Po activities in Mombasa creeks can be categorized as higher owing to the increase in the concentrations of this radionuclide in sediment compared to levels reported in other countries under similar anthropogenic influence. Whereas ^{210}Po in sediment from Mtwapa, Makupa and Tudor creek may seem low in activity, many marine organisms can bioaccumulate it in their tissues resulting in adverse effects (Carvalho and Fowler, 1993). More studies are therefore needed to determine bioaccumulation of ^{210}Po in biota and its potential effects, especially if such biota are consumed by human.

CRediT authorship contribution statement

Okuku Eric Ochieng: Conceptualization, Data curation, Funding acquisition, Investigation, Methodology, Project administration, Supervision, Validation, Writing - original draft, Writing - review & editing.**Kiteresi Linet Imbayi:**Data curation, Formal analysis,

Investigation, Methodology, Validation, Writing - original draft, Writing - review & editing.**Wanjeri Veronica Ogolla:**Conceptualization, Data curation, Formal analysis, Funding acquisition, Investigation, Methodology, Project administration, Resources, Software, Supervision, Validation, Visualization, Writing - original draft, Writing - review & editing.**Owato Gilbert Omondi:** Conceptualization, Data curation, Formal analysis, Funding acquisition, Investigation, Methodology, Project administration, Resources, Software, Supervision, Validation, Visualization, Writing - original draft, Writing - review & editing.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.marpolbul.2020.111040.

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