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Radioactivity Distribution In Surface And Core Sediment Of The Central Part Of The Algerian Coast: An Estimation Of The Recent Sedimentation Rate

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Abstract

Sediment core samples and marine surface sediments of identical composition, mostly muddy, were collected using a Van Veen type grab and a box corer on board the M.S. Benyahia research vessel (ISMAL), along the Algerian littoral, between Algiers (36° 49.9 N/ 03° 02.3 E) and Cherchell (36° 39.4 N/ 02° 12.4 E), during a sampling cruise in September 1997. The samples were analysed to determine the activity concentration of natural radionuclides (uranium and thorium series and ⁴⁰K as well) and artificial radionuclides (¹³⁷Cs and Pu isotopes), using a direct gamma spectrometry for gamma emitters and radiochemical separations and alpha spectrometry for alpha emitters. The horizontal and vertical distribution of the examined radionuclides were studied in the surface and core samples and an effort to estimate the sedimentation rate was attempted. The measured values range was: 17 - 26 Bq/Kg dry for uranium series radioisotopes, 18 - 32 Bq/Kg dry for thorium series radioisotopes, 311 - 690 Bq/Kg dry for ⁴⁰K, 0.4 - 11 Bq/Kg dry, for ¹³⁷Cs and 0.4 - 1.0 Bq/Kg dry for ²³⁹ + ²⁴⁰Pu.

Keyword: Marine environment, Radioactivity, Sedimentation, ¹³⁷Cs, Monitoring.

Introduction

The sources of radioactivity in the marine environment come mainly from atmospheric nuclear tests, discharges from nuclear and reprocessing plants, and accidental releases.

Among the major long-lived radionuclides already present or introduced into seas, the most important are: the radionuclides of uranium, thorium series, and ⁴⁰K for natural radioactivity, ¹³⁷Cs, ⁹⁰Sr and ²³⁹+²⁴⁰Pu for

artificial radioactivity. These, depending on their biogeochemical properties, are transported from catchment areas to the marine medium and then undergo several complex natural processes, depending on quantity of particles, marine circulation and resuspension of sediments (IAEA, 1988). The distribution of both natural and artificial radionuclides within the seabed can provide information on transient and vertical sediment

movement, accumulation and global radioactive inventory.

Within the framework of the marine research programme for the Algerian littoral of the Mediterranean Sea, surface sediment and core samples collected from the central part of the Algerian coast were analysed to determine activity in Bq/kg dry weight of ^{137}Cs , $^{239+240}\text{Pu}$, ^{40}K and uranium and thorium series, in order to study the distribution of both natural and artificial radionuclides within the seabed which can provide information on transient and vertical sediment movement, accumulation and global radioactive inventory.

Materials and Methods

Sampling

Core sediment (0-20 cm) and surface sediment samples (0-5 cm), were collected from the central part of the Algerian littoral between Algiers ($36^\circ 49.9\text{ N}/03^\circ 02.3\text{ E}$) and Cherchell ($36^\circ 39.4\text{ N}/02^\circ 12.4\text{ E}$) at a distance of 100 km. by means of a Van Veen-type grab and a box corer. They were sampled during September 1997, on board the *M.S. Benyahia* research vessel (ISMAL). Among the bulk of samples, 0.5 surface sediments were collected through a transient at different seawater depths ranging from 30m to 350m, and a core sediment sample was collected by means of a box corer at a depth of 350m and sectioned into 17 sub-samples. Locations of the sampling sites

are given in Table 1 and Fig.1. Upon collection, box cores were sectioned immediately on board in layers of 1 to 2 cm thickness and each sediment slice was placed in a plastic bag and weighed.

In the shore-based laboratory, the water content of each sediment sample was determined by weight loss after drying at 110°C until a constant weight was obtained.

The samples were labelled and brought to the laboratory, dried at 100°C , crushed, homogenised and stored for more than 20 days in 500cc polyethylene radon impermeable Marinelli beakers in order to obtain secular equilibrium amongst ^{226}Ra and its short-lived decay products.

These samples have undergone direct counting by gamma spectrometry and radiochemical separations followed by alpha spectrometry.

Radioactivity measurement

Gamma analysis

Fine grained sediments, collected at the different locations, were analysed by gamma spectrometry using a high purity Germanium detector of relative efficiency of 23% and resolution (FWHM) 1.8 KeV at 1332 KeV g-energy of ^{60}Co . A computerized multichannel analyser (4096 channels), with a Genie-PC software, was used to collect and analyse the spectra.

Table 1
Location of sampling stations.

Station N°	Latitude	Longitude depth (m)	Water	Sampling device
08	/	/	250m	B.C.*
09	$36^\circ 41.90'\text{ N}$	$002^\circ 46.00\text{ E}$	20m	V.V.G.*
10	$36^\circ 44.60'\text{ N}$	$002^\circ 45.90\text{ E}$	70m	B.C.
11	$36^\circ 47.30'\text{ N}$	$002^\circ 45.90\text{ E}$	100m	V.V.G.
12	$36^\circ 48.20'\text{ N}$	$002^\circ 46.10\text{ E}$	200m	V.V.G.
13	$36^\circ 50.90'\text{ N}$	$002^\circ 45.70\text{ E}$	350m	B.C.
14	$36^\circ 53.40'\text{ N}$	$002^\circ 46.20\text{ E}$	500m	V.V.G.
*B.C. : Box Corer				
*V.V.G. : Van Veen Grab				

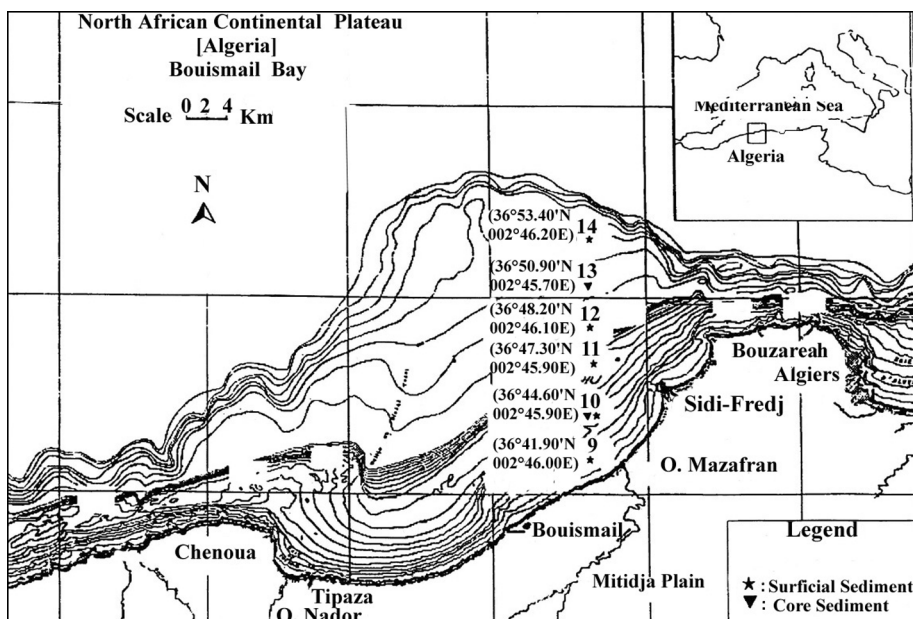


Fig. 1: Location of the sampling sites.

All samples were placed in a 500cm³ Marinelli beaker, put in contact with the detector and counted to give statistically reliable results (usually 16h - 24h). Detection efficiency of the system, for the analysed samples, was determined using a simulated standard source of ¹⁵²Eu of 11,655 Bq supplied by IAEA.

The concentrations of ¹³⁷Cs, ⁴⁰K, ²³⁸U and ²³²Th daughters were determined by using the gamma-rays of the highest emission probability of each radionuclide and also on the basis of secular equilibrium of uranium daughters. The ²²⁶Ra activity determination was based on ²¹⁴Pb (351.9 keV) and ²¹⁴Bi (609.3 keV), that of ²²⁸Ra from ²²⁸Ac (911.1 keV) and that of ²²⁸Th on ²¹²Pb (238.6 keV). The reliability and the accuracy of the method were checked by analysing reference materials provided by IAEA (IAEA - 384, IAEA - 326/327).

Alpha analysis

An approximate mass of 10g each sample was gradually ignited in a porcelain crucible in

a furnace up to 600 °C for 3h and held at that temperature for 6 hours to burn off the carbonaceous material. After ignition, samples were weighed in a Teflon beaker and wetted with about 10ml of 2 M HNO₃ to produce a slurry. With the aim of activity determination, an appropriate amount of ²³⁶Pu tracer in the range of 0.02 - 0.3 Bq was added to each sample.

The acid decomposition of the samples was succeeded by adding 40% HF acid to promote dissolution and removal of silica and 65% HNO₃ to remove as much HF and SiF₄ as possible and to convert the insoluble fluoride salts to soluble nitrate salts (Larosa, 1990 a). Plutonium and thorium separation was based upon the reduction-oxidation reaction produced by the addition of 100% hydrazinium hydroxide (N₂H₅OH) which converts all plutonium valences to the tetravalent and trivalent oxidation state (Larosa, 1990 a).

Before passing the sample solution onto a conditioned Bio Rad AG 1 x 4, 100 - 200 mesh anion exchange column, 5g of Na NO₂ was

added to convert any Pu (III) to Pu (IV). The Pu (IV) and Th (IV) are strongly sorbed onto the resin as anionic nitrate complexes, while the others (Am, U, Fe rare earth) are not.

Plutonium and thorium isotopes were successively eluted by passing 32% HCl and then 0.1 M NH₄I – 9 M HCl (Larosa, 1992).

Finally, plutonium and thorium isotopes were coprecipitated with NdF₃ by the addition of 5ml of 40% HF and the solution was dripped on a 0.1 µm pore size membrane filter (Ø d: 20 mm).

After washing with 5ml of 4% HF and 5ml of 20% ethanol and drying the filtered precipitate at about 50° C, the dried filter was carefully mounted onto a stainless steel disc to be counted by alpha spectrometry using a silicon surface barrier detector (Canberra series) and a multichannel analyser system.

Results and Discussion

The results of natural and man-made radionuclides measured by direct gamma

Table 2
Concentration in Bq kg⁻¹ dry weight of ¹³⁷Cs, ⁴⁰K, ²³⁸U and ²³²Th daughters in surface marine sediments* of the central part of the Algerian coast along the transect.

Sampling point (depth)	²²⁶ Ra	²¹² Pb	²²⁸ Ac	⁴⁰ K	¹³⁷ Cs
09 (20m)	17.32 ± 0.88	19.17 ± 0.87	18.46 ± 0.94	311.7 ± 14.6	0.88 ± 0.04
10 (70m)	19.35 ± 0.98	25.86 ± 1.17	26.46 ± 1.35	447.3 ± 21.0	3.28 ± 0.13
12 (200m)	22.48 ± 1.15	33.65 ± 1.53	28.36 ± 1.45	590.4 ± 24.7	10.96 ± 0.44
13 (350m)	22.87 ± 1.26	32.25 ± 1.47	25.88 ± 1.32	580.6 ± 23.4	5.75 ± 0.23
14 (500m)	26.15 ± 1.34	31.87 ± 1.45	31.05 ± 1.58	613.4 ± 25.5	5.82 ± 0.23

*Nature of sediment: Mud

Table 3
Concentration in Bq kg⁻¹ dry weight of ²³⁹⁺²⁴⁰Pu, ¹³⁷Cs, ⁴⁰K, ²³⁸U and ²³²Th daughters in core marine sediments * of the central part of the Algerian coast along the transect.

Samp N°	Depth (cm)	Dens. g/m ³	²²⁶ Ra	²²⁸ Ac (²²⁸ Ra)	²¹² Pb(²³² Th)	²¹⁰ Pb	⁴⁰ K	¹³⁷ Cs	²³⁹⁺²⁴⁰ Pu
1	0.5	1.18	22.9 ± 1.2	25.9 ± 1.3	32.3 ± 1.5	3.5 ± 1.2	581 ± 23	5.75 ± 0.2	
2	2	0.54	24.1 ± 1.2	26.3 ± 1.3	32.7 ± 1.5	3.2 ± 1.1	583 ± 23	6.29 ± 0.3	0.68 ± 0.06
3	3.5	0.52	24 ± 1.2	25 ± 1.3	27.9 ± 1.3	3.7 ± 1.3	603 ± 24	5.70 ± 0.2	
4	4.5	0.82	25.2 ± 1.3	26.7 ± 1.4	33.8 ± 1.5	3.3 ± 0.03	690 ± 24	5.6 ± 0.2	
5	5.5	0.87	21.8 ± 1.1	25.4 ± 1.3	27.9 ± 1.3	2.3 ± 1.1	604 ± 24.2	4.98 ± 0.2	1.02 ± 0.09
6	6.5	0.77	24.6 ± 1.3	28.6 ± 1.5	29.7 ± 1.4	2.3 ± 1.0	576 ± 23	5.23 ± 0.2	
7	7.5	0.85	23.9 ± 1.2	29.9 ± 1.5	31.5 ± 1.4	2.4 ± 1.1	609 ± 24.4	5.0 ± 0.2	
8	8.5	0.78	22.3 ± 1.1	31.3 ± 1.6	31.7 ± 1.5	1.8 ± 1.2	606 ± 24	4.65 ± 0.2	
9	9.5	0.88	22.5 ± 1.1	31.0 ± 1.6	32.1 ± 1.5	2.66 ± 1.2	592 ± 24	4.2 ± 0.2	0.72 ± 0.06
10	10.5	0.82	23.0 ± 1.2	29.0 ± 1.5	33.0 ± 1.5	1.8 ± 1.0	584 ± 23	3.3 ± 0.1	
11	11.5	0.83	22.7 ± 1.1	21.3 ± 1.1	30.3 ± 1.4	2.4 ± 1.0	585 ± 23	2.6 ± 0.1	
12	12.5	0.84	22.0 ± 1.1	30.0 ± 1.5	27.8 ± 1.3	0.7 ± 0.5	585 ± 23	2.1 ± 0.09	
13	13.5	0.89	22.0 ± 1.1	30.6 ± 1.6	32.7 ± 1.5	1.1 ± 0.5	573 ± 23	1.4 ± 0.06	
14	14.5	0.89	21.4 ± 1.1	29.4 ± 1.5	32.0 ± 1.5	1.0 ± 0.3	568 ± 23	1.2 ± 0.05	
15	15.5	0.94	23.0 ± 1.2	34.4 ± 1.8	35.2 ± 1.6	1.0 ± 0.01	615 ± 25	0.6 ± 0.02	0.16 ± 0.02
16	16.5	0.97	24.5 ± 1.2	32.6 ± 1.7	36.5 ± 1.7	0.73 ± 0.5	633 ± 25	0.4 ± 0.02	
17	18	0.72	23.4 ± 1.2	32.1 ± 1.6	33 ± 1.5	0.6 ± 0.4	623 ± 25	0.1 ± 0.004	
MDA	----	----	0.19	0.31	0.17	----	3.48	0.047	0.1 ± 0.01

spectrometry and alpha spectrometry in the analysed sediment samples are given in Table 2 and Table 3, which represent the concentrations of surface sediment along a transect and core sediment, respectively. The natural radionuclides consisted of ^{238}U and ^{232}Th daughters and ^{40}K . The detection limit of gamma emitters ranges from 0.5 to 3.5 Bq/kg dry. Concentration of ^{226}Ra was determined on the basis of the average value of ^{214}Pb and ^{214}Bi activity. Natural radioactivity present in the surface marine sediment along the transect (see Fig.1) varies from 17 to 26 Bq/kg dry for ^{226}Ra , from 18 to 32 Bq/kg dry for (^{212}Pb , ^{228}Ac) which seems to be in equilibrium, and from 312 to 613 Bq/kg dry for ^{40}K . Regarding the core sediment, ^{226}Ra concentration is more or less constant and is stabilised around an average value of 23.1 Bq/kg dry, ^{228}Ac and ^{212}Pb present an average value of 28.8 and 31.8 Bq/kg dry respectively with low variation, ^{40}K has an average concentration of 600.6 Bq/kg dry also with low variation.

As for artificial radionuclides, ^{137}Cs and $^{239+240}\text{Pu}$ were identified and measured at low levels. The detection limit of Pu isotope is around 0.1 Bq/kg dry and that of ^{137}Cs is about 0.5 Bq/kg dry. ^{137}Cs activity decreases from the top to bottom of the core, except in the recorded peak. Concerning surface sediment along the transect, we observe an increase of radioactivity from shore to offshore for ^{226}Ra , ^{212}Pb , ^{228}Ac and ^{40}K and also for ^{137}Cs . All radionuclides measured show a peak at station 12. Variation of activity concentrations in this transect is related to the transport of particles depending on the grain size and also to their distribution. Correlation between ^{40}K and ^{226}Ra is presented in Fig. 2. In this study, we can see a transient and vertical distribution of radioactivity. The most important aspect we can observe is the profile of ^{137}Cs and ^{210}Pb in the core sediment. Variation of activity of both radionuclides are plotted versus depth, as shown in Fig. 3 and Fig. 4. For ^{137}Cs , the peaks of 1963 and 1986 are observed at 5.5 and 2 cm respectively. The 1958 peak is not clear as this

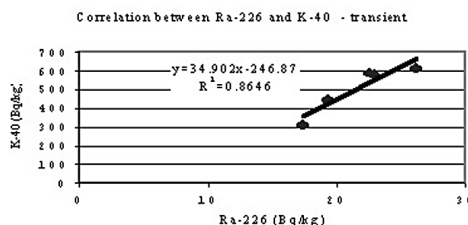


Fig. 2: Correlation between ^{40}K and ^{226}Ra , along the transect.

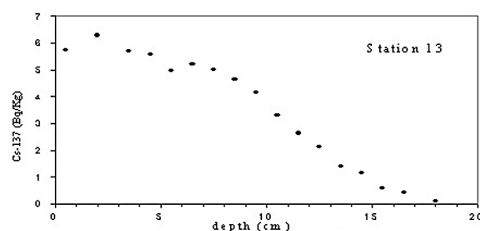


Fig. 3: Concentration variation of ^{137}Cs versus depth in the core sediment.

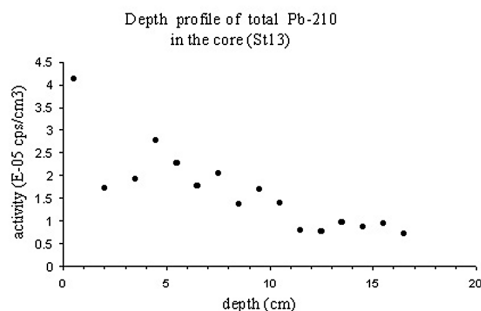


Fig. 4: Concentration variation of ^{210}Pb versus depth in the core sediment.

was initially a smaller scale input than the 1963 fallout and is then reduced by radioactive decay and sediment mixing (Cundy and Croudace, 1995), (Cochran, 1985). Concerning Pu isotopes, samples at depths of 2, 5, 9 and 15 cm were analysed in order to extract $^{239+240}\text{Pu}$. The measured concentrations range from 0.16 to 1.02 Bq/kg dry. Concentration ratios of ^{137}Cs to $^{239+240}\text{Pu}$ calculated at the above mentioned depths are 9.3, 4.9, 5.8 and 3.9 respectively. These values are close to those already found in the Algerian littoral (Nouredine, 1997). They are more or less constant over all the samples, and lower than those evaluated after the Chernobyl period (PNUE/AIEA, 1992).

A sedimentation rate of 0.18 cm/y is obtained from both peaks. Total ^{210}Pb profile in the core shows the expected exponential decrease with depth. A maximum value is observed near the sediment surface. At depths below 12 cm, ^{210}Pb activities decrease to comparatively constant values which correspond to the background levels of its parent ^{226}Ra .

Regarding ^{228}Th and ^{228}Ra profiles, the $^{228}\text{Th}/^{228}\text{Ra}$ activity ratio is about 1.25 at the top of the core and about 0.95 at 12 cm. Below this depth, equilibrium is reached between ^{228}Th and ^{228}Ra , with an activity ratio of around 1.0.

Conclusions

Regarding the obtained results, the concentrations of radionuclides in surface sediments appears to be correlated to the grain size.

As for the measured radionuclides of uranium, thorium series and ^{40}K , the concentrations in surface sediments are found to increase from shore (20m) to offshore (500m). The concentrations of ^{137}Cs and ^{210}Pb present a peak at a bottom depth of 200m.

In the core sediment samples, the natural radioisotopes showed constant values at any depth of the core, except for ^{210}Pb , which shows an exponential decrease with depth. Concerning ^{137}Cs and $^{239+240}\text{Pu}$ depth profiles, two subsurface maxima were observed and seem to be attributed to the Chernobyl accident and worldwide fallout peaks, respectively.

The deposition of ^{137}Cs in sediments at the coring location was calculated to be 511 Bq/m² by summing the concentrations of ^{137}Cs in the different core depths. The mean sedimentation rate obtained from both ^{210}Pb in excess and ^{137}Cs profiles was 0.18 cm/y.

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