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Radioactivity in Marine Salts and Sediments

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1. Introduction

Radioactivity is a natural phenomenon always taking place in our planet and in the whole universe. In the very beginning of matter, which it is evolving till now, some radioactive isotopes were created, among others, to form either in a mixture or as a single one, the ninety material units known as elements, which combined in a huge number of ways represent what is called matter, nature and universe. This sort of radioisotopes are, for example, ⁴⁰K, ⁵⁰V and ⁸⁷Rb, as well as every radioisotope found from bismuth to uranium, all of them radioactive, classified by Mendeleieff according their atomic number and weight in the Periodic Chart. These natural radioisotopes are called Primordial and are shown in Table 1.

Radioisotope	Half Life (years)	Isotopic Abundance (%)
⁴⁰ K	1.3x10 ⁹	0.0118
⁵⁰ V	6x10 ¹⁵	0.24
⁸⁷ Rb	4.7x10 ¹⁰	27.85
¹¹³ Cd	9x10 ¹⁰	12.3
¹¹⁵ In	6x10 ¹⁴	95.72
¹²³ Te	1.24x10 ¹³	0.87
¹³⁸ La	1.3x10 ¹¹	0.089
¹⁴⁴ Nd	2.1x10 ¹⁵	23.85
¹⁴⁷ Sm	1.1x10 ¹¹	15
¹⁴⁸ Sm	7x10 ¹⁵	11.2
¹⁵² Gd	1.1x10 ¹⁴	0.20
¹⁵⁶ Dy	2x10 ¹⁴	0.06
¹⁷⁶ Lu	3x10 ¹⁰	2.6
¹⁷⁴ Hf	2x10 ¹⁵	0.18
¹⁸⁷ Re	5x10 ¹⁰	62.6
¹⁸⁶ Os	2x10 ¹⁵	1.6
¹⁹⁰ Pt	6x10 ¹¹	0.0127
²⁰⁹ Bi	> 2x10 ¹⁸	100

Table 1. Radioisotopes in the isotopic mixture of elements from K to Bi (Primordial)
(Choppin a, 1980)

Two vacancies are shown in the Periodic Chart: Tc and Pm, elements not present in nature, because when they are produced by nuclear reactions, only short half life radioisotopes are produced, and so, if they have existed some time, they were quickly transformed into their neighbour elements. But nuclear reactions are taking place continuously in the earthly atmosphere by the interaction between light elements in gaseous state and nuclear particles such as α particles, fast neutrons, protons and deuterons coming from stratosphere. The products of these nuclear reactions are also radioisotopes, which are pulled down to the planet mainly by rain water and wind with no interruption. Radioisotopes of this sort are, for example: ^3H , ^{10}Be and ^{14}C , which in spite of their short half lives, compared with the age of solar system, reach an equilibrium state between their rates of production and decaying. These natural radioisotopes are called Cosmogenic and are shown in Table 2.

Radiosotope	Half Life	Production rate in the atmosphere (nucleus/ m ² -s)
^3H	12.35 years	2500
^7Be	53.4 days	81
^{10}Be	1.6×10^6 years	360
^{14}C	5715 years	22000
^{22}Na	2.6 years	0.6
^{26}Al	7.16×10^6 years	1.7
^{32}Si	280 years	
^{32}P	14.3 days	
^{33}P	25.3 days	
^{35}S	87.5 days	14
^{36}Cl	3×10^5 years	11
^{39}Ar	269 years	

Table 2. Some radioisotopes found in rain water (Cosmogenic) (Choppin b, 1980)

This is a very general and rather schematic description of natural radioactivity, always existent and main indicator of earth and universe evolution, since intensity of every radioactive source is always decreasing as time goes by, that is to say, the number of nucleus decaying by unit time when emitting nuclear radiations is inversely proportional to half life, and directly proportional to mass of every radioisotope, either natural or by human creation. But over the unavoidable and omnipresent natural radioactivity, it has been added that created by man. First radioisotopes of short half life were created, such as ^{13}N and ^{30}P with half lives of 9.9 and 2.5 minutes respectively, by the irradiation of B and Al with α particles emitted by Po. This discovery was made by Frederic Joliot and his wife Irene Curie in 1934. Since then, more than 2,000 artificial radioisotopes have been created, either as a research field itself or by a huge number of technological applications.

1.1 The Oklo phenomenon, a nuclear reactor in nature

In 1972, one mine of uranium minerals called Oklo, situated in the young country of Gabon, in Western Africa, was being fully exploited. Its minerals were sent to Pierrelate Centre for industrial uranium enrichment in France. Surprisingly, some samples showed a lower ^{235}U concentration than elsewhere in the world, that is to say 0.7%, which in some cases

decreased as much as 0.4%. An explanation of that anomaly was that some mass of ^{235}U had suffered fission for some time in the past. Residues of fission products with longer half-life were thus looked for in the site, and were surprisingly found. The minimum concentration of highly fissionable ^{235}U to have the critical mass for fission chain reaction is 1%. As this radioactive isotope half-life is 700 million years, while that of ^{238}U is 4.5 billion years, the necessary time span to get that minimum concentration finished 400 million years ago. But age of fission products' residues found in the field were also coincident within a much larger time span, around 2 billion years ago, which is also a common order of magnitude for some other minerals with some radioactive isotope in its composition, such as ^{40}K and ^{87}Rb . At that time, ^{235}U concentration in minerals should have been much greater, and thus very likely to make fission possible. Geographical conditions are favourable as well as rain may have washed out uranium minerals found in the surrounding hills, which could have then concentrated at the bottom of a lake. This lake could have then dried out as a result of a change in the rain cycle, or possibly as a consequence of fission heat, from which sediments can be found at Oklo mine. Therefore, the Oklo phenomenon is a fact that supports the idea of radioactivity as a natural component of material reality, and should by no means cause major concerns if the phenomenon is adequately managed, as happens with fire, explosives, acids, fuels, speed, pressure, electricity and so on. As Chang, the great chemist says: "humans are not necessarily the innovators, but merely the imitators of nature" (Chang, 2005).

Finally, some natural radioisotopes with comparable half life to planet age, such as heavy ^{232}Th , ^{235}U and ^{238}U are decaying into radioisotopes which linking one to another make a radioactive chain, each link created by decaying of the previous one and evolving to next one by its own decaying, to finish with a stable Pb isotope. These sort of natural radioisotopes are called Radiogenic and are shown in Tables 3, 4 and 5. So, they have as a link, for example, ^{215}At and ^{218}At , radioisotopes with extremely short half lives, but in spite of it always present in nature because they are continuously created in the ^{235}U and ^{238}U radioactive chains. Pu radioisotopes are formed by ^{238}U irradiated with thermal neutrons and successive beta decay. Among them, ^{239}Pu ($t_{1/2} = 24,400$ years) and ^{241}Pu ($t_{1/2} = 13.2$ years) are the most important, because they have a great cross section for fission with thermal neutrons, and so they are the origin of the so called breeding reactors, where calorific energy is obtained at same time that a new fissionable, nuclear fuel is produced.

^{232}Th radioactive chain is called (4n) because the mass number of every link is a multiple of 4. In the same way, as radioactive chains of ^{238}U and ^{235}U show links whose mass numbers are reproduced by algebraic expressions (4n+2) and (4n+3), where n is an entire number, they are called in this manner. While ^{241}Pu radioactive chain, which is not natural, but produced in modern enriched uranium nuclear reactors, is called (4n+1) by same reason. It is noticeable from Tables 3, 4 and 5 the presence of links with Ra, Rn and Po isotopes. Ra and Po were the first radioisotopes isolated from pechblenda minerals by Pierre and Marie Curie, while Rn radioisotopes are also found there, all of them with different half lives and radiation energies. These radioisotopes of heaviest noble gas have been always a radioactive component of earth atmosphere everywhere, specially concentrated in those indoor places where their α and γ radiations are now detected. Therefore, emissions produced by natural radioisotopes have always been in air, earth and sea, but quite a different matter is the environmental contamination produced today by ^{235}U and maybe tomorrow by ^{239}Pu and ^{241}Pu fission products.

Radioisotope	Half Life	Historical Name	Type of radioactive decay
^{232}Th ↓	1.4×10^{10} years	Thorium	α
^{228}Ra ↓	6.7 years	Mesothorium I	β^-
^{228}Ac ↓	6.13 hours	Mesothorium II	β^-
^{228}Th ↓	1.9 years	Radiothorium	α, γ
^{224}Ra ↓	3.64 days	Thorium X	α, γ
^{220}Rn ↓	55 seconds	Toron (emanation)	α, γ
^{216}Po ↓	0.15 seconds	Thorium A	α
^{212}Pb ↓	10.6 hours	Thorium B	β^-, γ
^{212}Bi ↓	60.6 minutes	Thorium C	α, β^-, γ
$^{212}\text{Po} (64\%)$ ↓	304 nanoseconds	Thorium C'	α
$^{208}\text{Tl} (36\%)$ ↓	3.1 minutes	Thorium C''	β^-, γ
^{208}Pb	stable	Thorium D	—

Table 3. ^{232}Th radioactive chain (4n)

Radioisotope	Half Life	Historical Name	Type of radioactive decay
^{238}U ↓	4.5×10^9 years	Uranium I	α
^{234}Th ↓	24.1 days	Uranium X ₁	β^-, γ
$^{234\text{m}}\text{Pa}$ ↓	1.17 minutes	Uranium X ₂	β^-, γ

Radioisotope	Half Life	Historical Name	Type of radioactive decay
^{234}Pa ↓	6.75 hours	Uranium Z	β^- , γ
^{234}U ↓	2.5×10^5 years	Uranium II	α , γ
^{230}Th ↓	8×10^4 years	Ionium	α , γ
^{226}Ra ↓	1602 years	Radium	α , γ
^{222}Rn ↓	3.8 days	Radon (emanation)	α , γ
^{218}Po ↓	3.05 minutes	Radium A	α , β^-
^{214}Pb (99.98%) ↓	26.8 microseconds	Radium B	β^- , γ
^{218}At (0.02%) ↓	2 seconds	Astatine	α
^{214}Bi ↓	19.7 minutes	Radium C	α , β^- , γ
^{214}Po (99.98%) ↓	164 microseconds	Radium C'	α , β^-
^{210}Tl (0.02%) ↓	1.3 minutes	Radium C''	β^- , γ
^{210}Pb ↓	21 years	Radium D	β^- , γ
^{210}Bi ↓	5.01 years	Radium E	α , β^-
^{210}Po (100%) ↓	138.4 days	Radium F	α
^{206}Tl (0.00013%) ↓	4.19 minutes	Radium E'	β^-
^{206}Pb	stable	Radium G	—

Table 4. ^{238}U radioactive chain ($4n + 2$)

Radioisotope	Half Life	Historical Name	Type of radioactive decay
^{235}U ↓	7.1×10^8 years	Actinouranium	α , γ
^{231}Th ↓	25.2 hours	Uranium Y	β^- , γ
^{231}Pa ↓	3.25×10^4 years	Protoactinium	α , γ
^{227}Ac ↓	21.6 years	Actinium	α , β^- , γ
$^{227}\text{Th}(98.6\%)$ ↓	18.2 days	Radioactinium	α , γ
$^{223}\text{Fr}(1.4\%)$ ↓	22 minutes	Actinium K	β^- , γ
^{223}Ra ↓	11.43 days	Actinium X	α , γ
^{219}Rn ↓	4 seconds	Actinium (emanation)	α , γ
^{215}Po ↓	1.8 milliseconds	Actinium A	α , β^-
$^{211}\text{Pb}(100\%)$ ↓	36.1 minutes	Actinium B	β^- , γ
$^{215}\text{At}(0.00023\%)$ ↓	0.1 millisecond	Astatine	α
^{211}Bi ↓	2.15 minutes	Actinium C	α , β^- , γ
$^{211}\text{Po}(0.28\%)$ ↓	0.52 seconds	Actinium C'	α , γ
$^{207}\text{Tl}(99.7\%)$ ↓	4.79 minutes	Actinium C''	β^- , γ
^{207}Pb	stable	Actinium D	—

Table 5. ^{235}U radioactive chain ($4n + 3$)

Radioisotope	Half Life	Name	Type of radioactive decay
^{241}Pu ↓	13.2 years	Plutonium	α , β^- , γ
$^{241}\text{Am}(100\%)$ ↓	458 years	Americium	α , γ
$^{237}\text{U}(0.0023\%)$ ↓	6.75 days	Uranium	β^- , γ
^{237}Np ↓	2.14×10^6 years	Neptunium	α , γ
^{233}Pa ↓	27 days	Protactinium	β^- , γ
^{233}U ↓	1.6×10^5 years	Uranium	α , γ
^{229}Th ↓	7340 years	Thorium	α , γ
^{225}Ra ↓	14.8 days	Radium	β^- , γ
^{225}Ac ↓	10 days	Actinium	α , γ
^{221}Fr ↓	4.8 minutes	Francium	α , γ
^{217}At ↓	0.032 seconds	Astatine	α
^{213}Bi ↓	47 minutes	Bismuth	α , β^- , γ
$^{213}\text{Po}(97.8\%)$ ↓	4.2 microseconds	Polonium	α
$^{209}\text{Tl}(2.2\%)$ ↓	2.2 minutes	Thallium	β^- , γ
^{209}Pb ↓	3.3 hours	Lead	β^- , γ
^{209}Bi	$> 2 \times 10^{18}$ years	Bismuth	α ?

Table 6. ^{241}Pu radioactive chain ($4n + 1$)

2. Radioactive contamination

Radioactive contamination started on the planet in 1945, when the first nuclear test was performed in Alamo Gordo, New Mexico, followed by the war actions in Hiroshima and Nagasaki. Since then, radioactive contamination at global level has been variable, depending on repeated nuclear tests, few accidents such as Three Mile Island and Chernobyl, and minor failures in nuclear power plants. These contaminants are produced mainly by fission products from ^{235}U , which according their fission yielding and half lives, they remain radioactive during a time span from seconds to a great number of eons ($1 \text{ eon} = 1 \times 10^9 \text{ years}$). But certainly, burned nuclear fuels which are under control and stored accordingly the safest techniques to guarantee they will always be confined and never disseminated in the environment, same case that residues of artificially produced radioisotopes used in medicine, industry or any other purpose, they should not be considered as radioactive contaminants, as much as they are under safe enough surveillance. So, approximately 30-40% all of known radioisotopes are fission products, which when they come into environment by deliberate nuclear explosion, severe accident or failure in nuclear plant, they represent the so called radioactive contamination. From this perspective, it seems that radioactive contamination has been growing up from its beginning, with rather short equilibrium periods. Also, if it is considered that sea water represents approximately 80% of planet surface, plus the action of wind, rain and rivers current, the main repository of radioactive contamination should be the sea. However, radioactive contamination is only added to natural radioactivity. From the first elements in the Periodic Table: ^3H , ^{10}Be and ^{14}C , natural radioisotopes are either continuously produced by nuclear reactions in the earthly atmosphere, or they were created at same time that non radioactive ones, in the mixture of isotopes forming elements such as ^{40}K , ^{50}V and ^{87}Rb . And then from Bi to beyond uranium elements, every isotope is radioactive with no exception. Therefore, it seems that to properly quantify the importance at planet level of any radioactive contamination, it should be done on the basis of radioactivity already present since the planet birth, whose decaying becomes the most evident sign of earth evolution and it is still taking place. In this way, 0.0118% isotopic abundance, $1.28 \times 10^9 \text{ years}$ half life, ^{40}K is the natural radioisotope most abundant in the earth crust and also in the numerous salts dissolved in sea water. So, the radioactivity due to ^{40}K might be the most suitable measurement, in order to have one basis of natural radioactivity to be compared with that of any artificial radioisotope. Among these, the fission product ^{137}Cs presents the highest yielding in the fission of ^{235}U , and it is the most common radioactive pollutant found in nuclear accidents due to its half life equal to 30.07 years, and γ rays easy to detect with higher efficiency due to a low energy equal to 662 Kev. Figure 1 represents the fission products yielding from ^{235}U vs. mass number (A) and Fig. 2 represents percentage of elements on earth vs atomic number (Z).

3. Experimental

3.1 Sampling and samples conditioning

Therefore, according with the idea to consider radioactivity as a quite natural phenomenon, supported by the existence of Primordial, Cosmogenic and Radiogenic radioisotopes, as well as the Oklo phenomenon, it is proposed to identify the natural radioactivity by Primordial radioisotope ^{40}K , based on the fact that it is present in one of more abundant elements on earth, as it can be seen in Fig. 2, and as a consequence is found in the

Log % ²³⁵U Fission Products Yielding

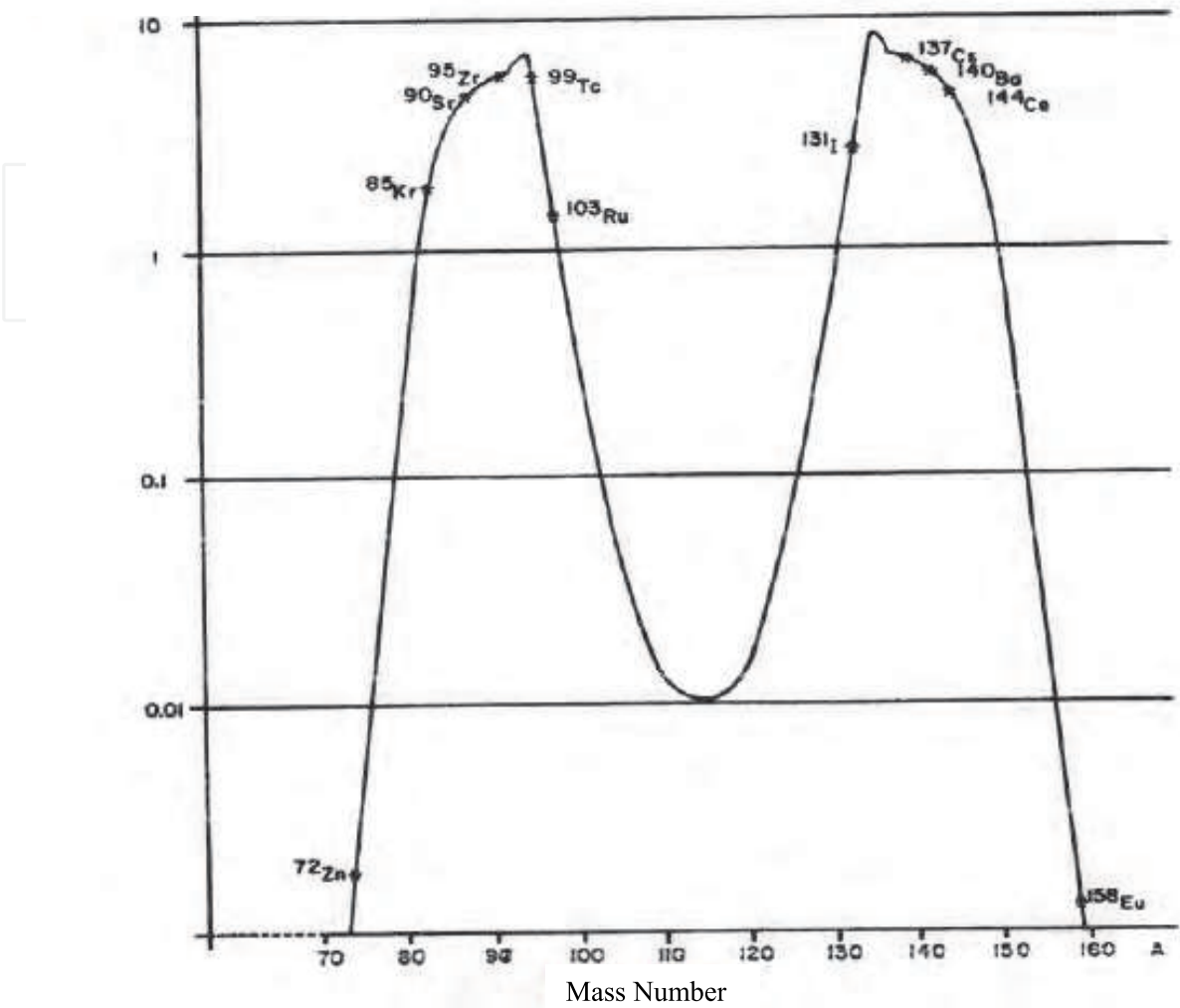


Fig. 1. ²³⁵U Fission Products Yielding vs. Mass Number (A)

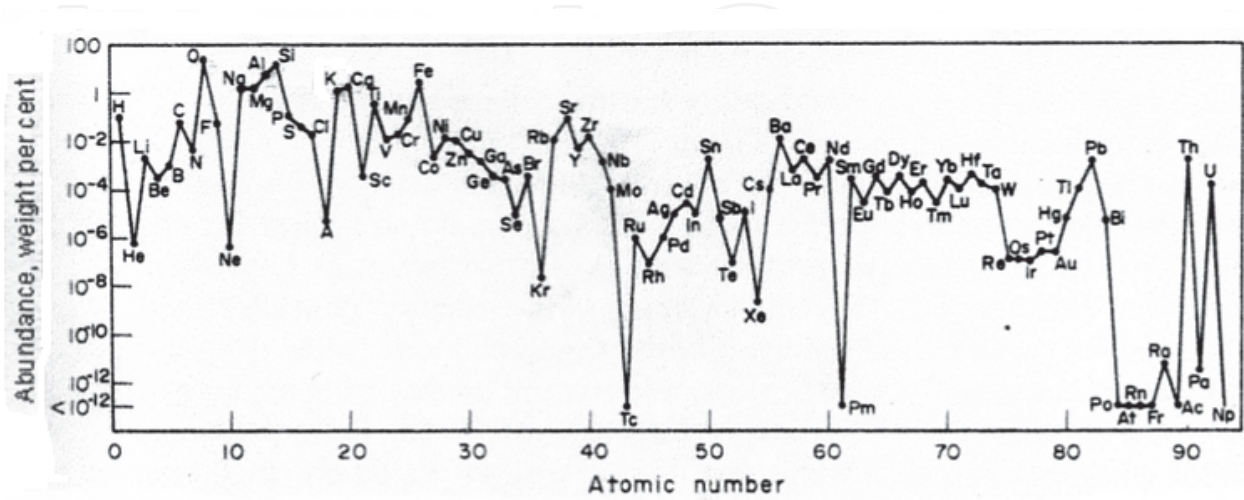


Fig. 2. Abundance of elements in earth (%) vs. Atomic Number (Choppin c, 1980)

radioactive background all over the world, while the present radioactive contamination can be easily represented by ^{137}Cs , fission product of ^{235}U . Besides, both radioisotopes are electromagnetic radiation emitters with suitable energies to be easily detected, and so one way to measure the intensity of present radioactive contamination should be to obtain a radioactive contamination factor (RCF), by dividing specific radioactivity of ^{40}K by that of ^{137}Cs in solid samples, that is to say disintegrations per time and weight units measured in both radioisotopes. This present radioactive contamination background, even when proceeds from limited portions on earth surface, where it has remained for long time as a well located radioactive source which must be left away by population and conveniently shielded, it has been unavoidable that a fraction of it spreads out to atmosphere in the gas and dust form, which can travel long distances to be finally carried down mainly by rain water on earth surface as either solutions or suspensions. But as sea represents the much larger proportion of planet surface, about 80%, and it is also the main factor of rain cycle, out of control radioactive pollutants produced anywhere in considerable amounts reach always the sea water in concentrations which can be easily measured by γ rays detection. Therefore, it seems that it is in sea water and marine sediments where global radioactive contamination should be searched and evaluated, because it is there where planet radioactive contamination has mainly created a growing deposit since the last world war. However, if it is assumed the sea water volume approximately as $1.4 \times 10^{18} \text{ m}^3$, then it might be considered as an enormous natural radioactive source, not at all by contamination, but because it contains in solution an important concentration of K salts and its natural radioisotope ^{40}K (β^- and γ rays emitter after electronic capture, half life 1.28×10^9 years, 0.0118% isotopic abundance), which represents the main source of natural radioactivity as much in solid minerals (excepting those of heavy metals from Pb on), as in sea water and marine sediments. In this way, in order to asses the importance of any present or future radioactive contamination at planet scale, it might be compared by some radioactive contamination factor or some other way with natural radioactivity, which has been increased at certain extent by radioactive contamination. We are talking here about radioactivity spread out to environment from a local point, which must be immediately attended in situ, whereas that diluted in environment and reaching far away places usually produces great panic, even when it has never before been compared with natural, already existent radioactivity since the beginning of solar system. On the other hand, ^{40}K radioactivity as well as K concentration salts in sea water increases with ocean depth till a maximum value, and then decreases before reaching the bottom till a value usually lower than that at surface, as it happens with every mineral salt dissolved in sea water (Vázquez, 2001). So, it is quite possible to characterize superficial sea water in different coasts in terms of ^{40}K specific radioactivity, by sampling at about one kilometre from the coast, where it keeps constant for parallel much longer distances on the littoral, and obviously is easier to do it that in high sea, useful figure to calculate the concentration of elementary K in that particular sea zone. The way to do it is quite simple: 6-8 litres of sea water must be boiled, in order to get a suitable volume of sea salt to fill up a Marinelli container, usually about half a litre, necessary to perform low background radioactive detection. Once the dry salt sample is weighed and conditioned in the Marinelli container, it is ready to measure its natural as well as polluting radioactivity, by making use first of one heavily shielded scintillation set (NaI, Tl activated), and then one equally shielded hyper-pure Ge detector (HPGe), during 12-

24 hours detection time. Also, sediment marine samples have been picked up from 40-60 meters depth in three zones: Gulf of Mexico, to south east of Veracruz port and Laguna Verde Nuclear Power Plant, around Grijalva and Usumacinta delta rivers, as well as north, near the border with territorial USA sea water, and in Pacific Ocean between Cortés sea and Mazatlán port. Samples were taken by two ships: Puma in the Gulf and Justo Sierra in the Pacific Ocean, both at service of Sea Science and Limnology Institute, from National University of Mexico. Figure 3 presents the Puma ship. Figure 4 the Justo Sierra ship. These ships work in Oceanography research, for Institute of Sea Science and Limnology, in the National University of Mexico. Figure 5 presents one sediment sample conditioned in the Marinelli container. Figure 6 presents the low background scintillation set and Figure 7 presents the low background semi-conductor set.



Fig. 3. Ship Puma, samples collector in Pacific Ocean



Fig. 4. Ship Justo Sierra, samples collector in Gulf of Mexico



Fig. 5. Marinelli container with sediments



Fig. 6. Scintillation Detection set

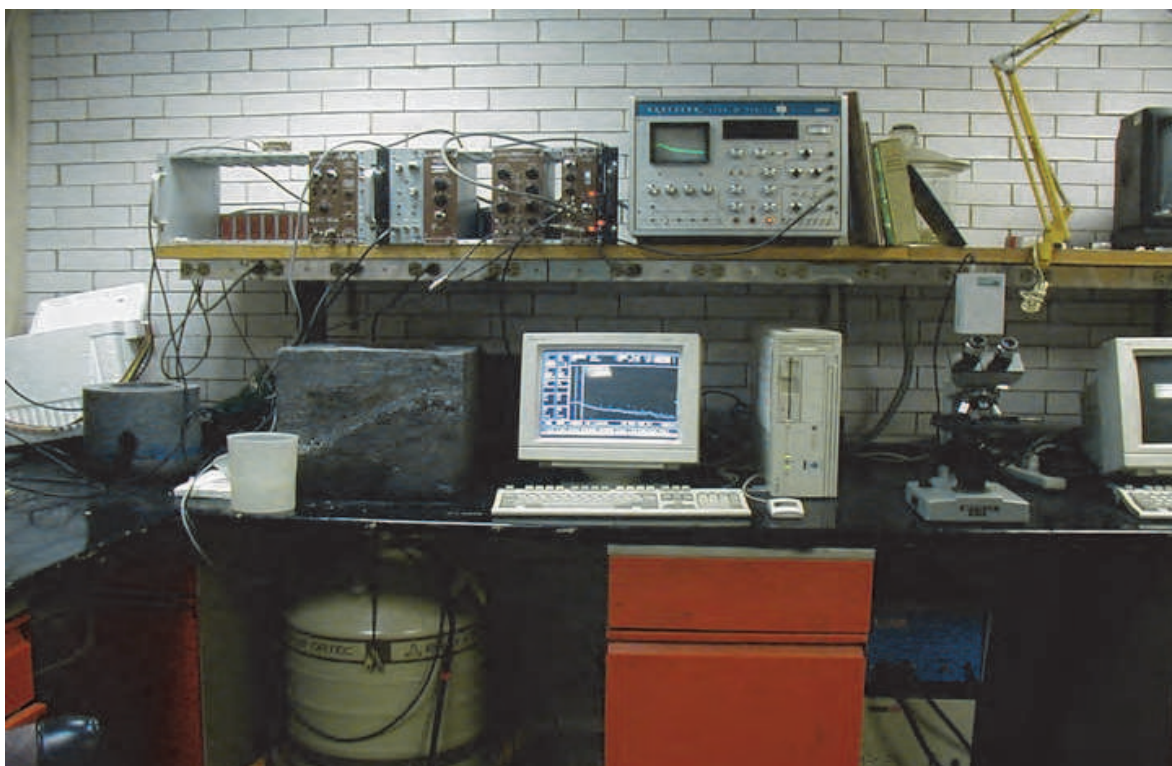


Fig. 7. HPGe Semiconductor Detection set

3.2 Radioactive detection

In order to obtain our results either of natural or contaminant radioactivity in Bq per gram of sea salts and marine sediments, we must calculate the detection efficiency of both, scintillation and HPGe detector systems. It is easier and more precise to use one ^{40}K calibrated source formed by a known weight of KCl, and by separate one ^{137}Cs calibrated source. Detection efficiency for the 1461 Kev γ rays peak emitted by ^{40}K was determined by a standard made out by filling a Marinelli container with a weighed mass of KCl salt, AR grade. Detection time of 10-20 minutes was enough to get $\pm 1\%$ as statistical error. Then, the counts accumulated in the peak expressed as counts per second (cps), when divided by the specific activity expressed as disintegrations per second per gram ($\text{dps/g} = \text{Bq/g}$) of either KCl or elementary K, and multiplied by 100, is obtained detection efficiency for scintillation and semiconductor systems in the same way. Equations 1 and 2 show the calculation to get the specific activity of KCl and elementary K respectively, due to 11% of ^{40}K decaying nucleus by electron capture to ^{40}Ar and emitting γ rays with an energy of 1461Kev. Equation 3 show the calculation to get the total specific activity of elementary K, due to 0.0118% isotopic abundance of ^{40}K (β^- emitter 89%, EC and γ rays emitter 11%), constant value that will be used to characterize sea salts.

$$\begin{aligned} \text{Bq } ^{40}\text{K} \rightarrow ^{40}\text{Ar} / \text{gKCl} &= 0.693 \times 6.02 \times 10^{23} \times 0.0118 \times 11 / 1.28 \times 10^9 \times 365 \times 24 \times 60 \times 60 \times 100 \times 100 \times 74.5 \\ &= 1.8 \text{ Bq } ^{40}\text{K} \rightarrow ^{40}\text{Ar} / \text{g KCl} \end{aligned} \quad (1)$$

$$\begin{aligned} \text{Bq } ^{40}\text{K} \rightarrow ^{40}\text{Ar} / \text{gK} &= 0.693 \times 6.02 \times 10^{23} \times 0.0118 \times 11 / 1.28 \times 10^9 \times 365 \times 24 \times 60 \times 60 \times 100 \times 100 \times 39.1 \\ &= 3.4 \text{ Bq } ^{40}\text{K} \rightarrow ^{40}\text{Ar} / \text{gK} \end{aligned} \quad (2)$$

$$\begin{aligned} \text{Bq } ^{40}\text{K} / \text{gK} &= 0.693 \times 6.02 \times 10^{23} \times 0.0118 / 1.28 \times 10^9 \times 365 \times 24 \times 365 \times 60 \times 60 \times 100 \times 39.1 \\ &= 31.19 \text{ Bq/gK} \end{aligned} \quad (3)$$

Where:

$$\text{Ln } 2 = 0.693$$

$$\text{Avogadro's number} = 6.02 \times 10^{23}$$

$$\text{Isotopic abundance of } ^{40}\text{K} = 0.0118/100$$

$$\text{Decay yielding of } ^{40}\text{K} \rightarrow ^{40}\text{Ar} = 11/100$$

$$\text{Half life of } ^{40}\text{K} = 1.28 \times 10^9 \text{ years} = 1.28 \times 10^9 \times 365 \times 24 \times 60 \times 60 \text{ seconds}$$

$$\text{KCl molecular weight} = 74.5$$

$$\text{K atomic weight} = 39.1$$

Therefore, detection efficiency for counts accumulated in either scintillation or semiconductor detector, produced by γ rays with energy 1461 Kev, emitted by ^{40}K , is given alternatively by equations 4 and 5.

$$\text{Det. Eff. (electromagnetic radiation) (\%)} = \text{cps} \times 100 / 1.8 \times W_{s1} \quad (4)$$

$$\text{Det. Eff. (electromagnetic radiation) (\%)} = \text{cps} \times 100 / 3.4 \times W_{s2} \quad (5)$$

Where:

cps = counts accumulated per second

1.8 = specific activity of $^{40}\text{K} \rightarrow ^{40}\text{Ar}$ by EC, γ rays emission per gram of KCl

($\text{Bq } ^{40}\text{K} \rightarrow ^{40}\text{Ar} / \text{g KCl}$)

3.4 = specific activity of $^{40}\text{K} \rightarrow ^{40}\text{Ar}$ by EC, γ rays emission per gram of elementary

K ($\text{Bq } ^{40}\text{K} \rightarrow ^{40}\text{Ar} / \text{g K}$)

W_{s1} = Weight of KCl in the Marinelli container

W_{s2} = Weight of K in the Marinelli container (52.48% of KCl)

In order to obtain the detection efficiency for gamma rays (662 Kev) emitted by radioactive contaminant ^{137}Cs , it has been used a calibrated multinuclide standard source in an identical Marinelli container to that used with KCl. In this case, calculation is only to divide counts per second accumulated in the corresponding peak (662 Kev), multiply by 100 and divide by the ^{137}Cs certificate activity in Bq at a given date and corrected to present time by decaying factor. To calculate detection efficiency by separate of γ rays from ^{40}K (1461 Kev) and γ rays from ^{137}Cs (662Kev), it is easier and more precise in our project, that to find that corresponding to ^{40}K from a graph efficiency versus energy, plotted with data obtained from the calibrated multinuclide source, because in this later case Compton distribution is much higher than in natural samples, such as KCl, marine salts and sediments. So, background correction in both detections has revealed as almost irrelevant when detections efficiencies are obtained, while on the other hand it is extremely important when marine salts and

sediments are detected during much longer time periods, from 20 to 24 hours, but with similar dead time in detectors to that produced by KCl source.

If samples from Oklo uranium mine were considered as marine sediments, in order to evaluate the radiation danger they represent, it is very likely that radioactivity from natural radioisotopes of heavy metals such as ^{232}Th , ^{235}U and ^{238}U , origin of radioactive chains with several short half life radioisotopes in their links, were substantially higher than that from ^{40}K , natural radioisotope present almost everywhere, and by sure in Oklo minerals too. Since also in marine sediments have been found radioactive heavy metals, similarity between these two mineral samples becomes more understandable, besides the hypotheses that Oklo mine was a huge lake, probably of salted water in its origin. So, even when radioactive contamination by ^{137}Cs is not possible to confirm in Oklo due to its relatively short half life, it should be very easily detected in marine salts in the case of recent contamination, such as that in Fukushima, Japan, which at present should be in the mixture of natural marine salts, and in the near future will be in marine sediments, accompanying heavy metals and of course ^{40}K .

3.3 Characterization of marine salts and sediments through natural and pollutant radioactivity

Samples were taken in two points of Gulf of Mexico. One is to the south east of Laguna Verde Nuclear Plant, between delta of Usumacinta and Grijalva rivers, and the other to the north east of the Gulf, near the line with territorial USA waters. In the Pacific Ocean, samples were taken from Cortés Sea to Mazatlán port. In order to characterize sea waters by its K concentration, 5-6 litres of water samples were boiled to obtain about half a Kilogram of salt to fill up one Marinelli container. The weight of salt obtained and divided by the number of litres evaporated gives us one first figure equal to g/L, which means salinity. When counts accumulated during 20-24 hours in a low background detection system, either scintillation or HPGe, are expressed as counts per second, corrected for background in same units (cps) and divided by salt sample weight, detection efficiency for 1461 Kev γ rays (2.8% in our scintillation system and 0.22% in our HPGe detector) and the fraction of ^{40}K nucleus decaying to ^{40}Ar by EC and γ rays emission (11/100), total specific activity of ^{40}K expressed as Bq/g of salt is obtained, according the equation 6:

$$\text{Bq/g salt} = (\text{cps}[\text{Sample}] - \text{cps}[\text{Background}]) / W_s \times \text{Det. Eff.} \times 0.11 \quad (6)$$

Where:

$\text{Bq/g salt} = \text{Specific activity of sea salt due to } ^{40}\text{K} \text{ total decaying } (\beta^- [89\%], \gamma \text{ rays } [11\%])$

$(\text{cps}[\text{sample}] - \text{cps}[\text{Background}]) = \text{counts accumulated per second by sample and corrected by background}$

$W_s = \text{Salt sample weight expressed in grams}$

$\text{Det. Eff} = \text{Detection efficiency for 1461 Kev } \gamma \text{ rays emitted by } ^{40}\text{K} \text{ in our detection systems, expressed as fractions } (\text{Scintillation} [2.8 \times 10^{-2}], \text{HPGe} [0.22 \times 10^{-2}])$

$0.11 = \text{Fraction of } ^{40}\text{K} \text{ nucleus decaying to } ^{40}\text{Ar} \text{ by EC and } \gamma \text{ rays emission } (11\%)$

In this way, when salinity is multiplied by specific activity of sea salt, activity per litre of sea water is obtained. Also, when specific activity of sea salt is divided by specific activity of

elementary K and multiplied by 100, concentration of K in sea salt is obtained as percentage, according the equations 7 and 8:

$$\text{Bq/L} = \text{g/L} \times \text{Bq/g salt} \quad (7)$$

$$\%K = \text{Bq/g salt} \times 100 / 31.19 \text{ Bq/g K} \quad (8)$$

Where:

Bq/L = Activity per litre of sea water due to ^{40}K total decaying (β^- [89%], γ rays [11%])

g/L = Salinity of sea water expressed in grams per litre of sea water

Bq/g salt = Specific activity of sea salt due to ^{40}K total decaying (β^- [89%], γ rays [11%])

$\%K$ = K concentration of sea salt expressed as percentage

$31.19 \text{ Bq}^{40}\text{K} / \text{g K}$ = Specific activity of elementary K due to ^{40}K total decaying (β^- [89%], γ rays [11%])

So, when these figures are experimentally obtained, a great portion of sea water may be characterized from the ^{40}K natural decaying of its salt, data which should be very useful to detect and evaluate any recent contamination, such as that occurred in Fukushima, Japan, at present, and in the past those of Three Miles Island in USA, and Chernobyl in Russia, even when the nuclear accident or failure might have occurred at a large distance from the sea site. In any case, radioactive contamination should be represented by some fission product, most probably ^{137}Cs , due to its high fission yielding and easy detection of 662 Kev γ rays emission. Nevertheless, and even when ^{137}Cs has not been detected in Mexican marine salts till now, it has been detected in every marine sediment tested in samples picked up at 60-80 meters deep. This fact maybe becomes enough evidence that it does already exists a radioactive contamination at sea bottom, creating one background from now on, which should be very important to evaluate in order to compare how it is growing up or maybe decaying when time goes by, and with no doubt nuclear power will have a great development all over the world. The main origin of this radioactive background at sea bottom, should be the test nuclear explosions at Alamo Gordo and Bikini, as well as the war actions in Hiroshima and Nagasaki, followed by nuclear test explosions performed by several countries since then, and only in a minor proportion by accidents and failure events of nuclear plants, considering that from 1945 to present day only 2.2 time spans of 30.07 years (half life of ^{137}Cs) have passed away. ^{137}Cs has not been detected so far in sea salt samples taken up from Mexican littorals, neither Pacific Ocean nor Gulf of Mexico. On the contrary, every sediment picked up from 60-80 meters depth, seems to have accumulated a small amount of ^{137}Cs , creating a certain pollutant radioactivity over the natural radioactive background present at sea bottom, which is represented mainly by ^{40}K and ^{232}Th , ^{235}U and ^{238}U radioactive chains. So, fission product ^{137}Cs should have been first dissolved in sea water, among a great diversity of ions in there, and then settled down on sediments as time goes by, because it is a rather heavy ion. In this way, ^{137}Cs present in sea salts should be indicating some recent pollution, while in marine sediments should be one of the main contributors to increase its natural background. Therefore, the proportion expressed as percentage of specific pollutant radioactivity $\text{Bq}^{137}\text{Cs/g}$ multiplied by 100 and divided by specific natural radioactivity ($\text{Bq}^{40}\text{K/g}$), should be as useful

in sea salts as in marine sediments, to have a reliable and easy to understand figure to evaluate the magnitude of recent pollution as well as to size up the possible growing or decreasing rate in already existing radioactive pollution in marine sediments.

4. Results

Figures 8 and 9 show the background and electromagnetic radiation (γ rays) of marine sediments picked up at Gulf of Mexico North, obtained with a low background scintillation detector, NaI(Tl), 3X3", coupled to a PC charged with Maestro Program.

Figures 10 and 11 show the background and electromagnetic radiation (γ rays) of marine sediments picked up at Gulf of Mexico North, obtained with a low background semiconductor detector, HPGe, coupled to a PC charged with Maestro Program II.

Table 7 shows the results obtained from sea salts samples taken up in Pacific Ocean North, between Cortes sea and Mazatlan port, and Gulf of Mexico North and South East, as well as sediments pollution measured by RCF (Radioactive Contamination Factor), where $RFC = \text{Bq } ^{137}\text{Cs} \times 100/\text{g} / \text{Bq } ^{40}\text{K}/\text{g}$.

These results have been obtained within statistical variations given by Maestro Program I and II, maximum ± 15 % to minimum ± 1% of counts accumulated in both detection systems during detection times from 3.96×10^4 to 8×10^4 seconds or 11 and 22.2 hours. So, when subtracting background and dividing activity due to ^{137}Cs by that due to ^{40}K , statistical variations were always below ± 15%.

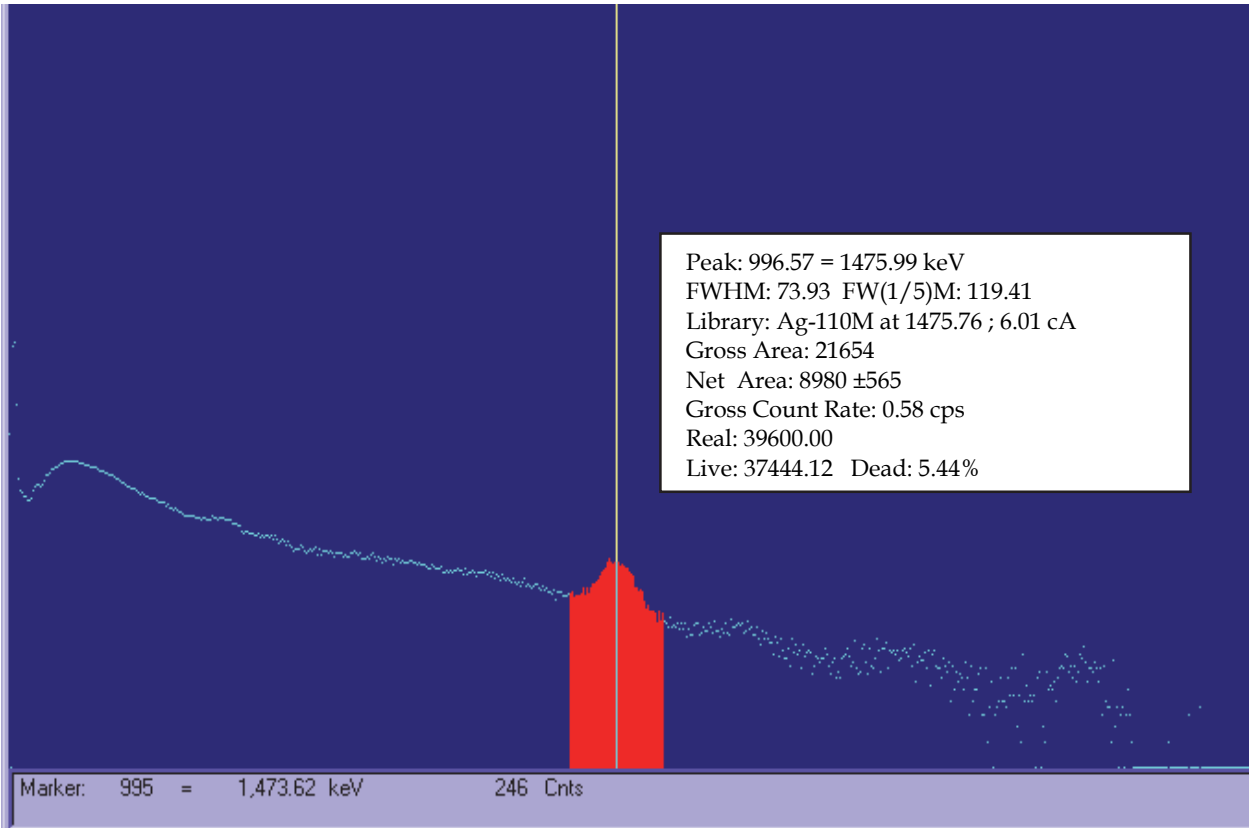


Fig. 8. Background spectrum in Scintillation Detection System

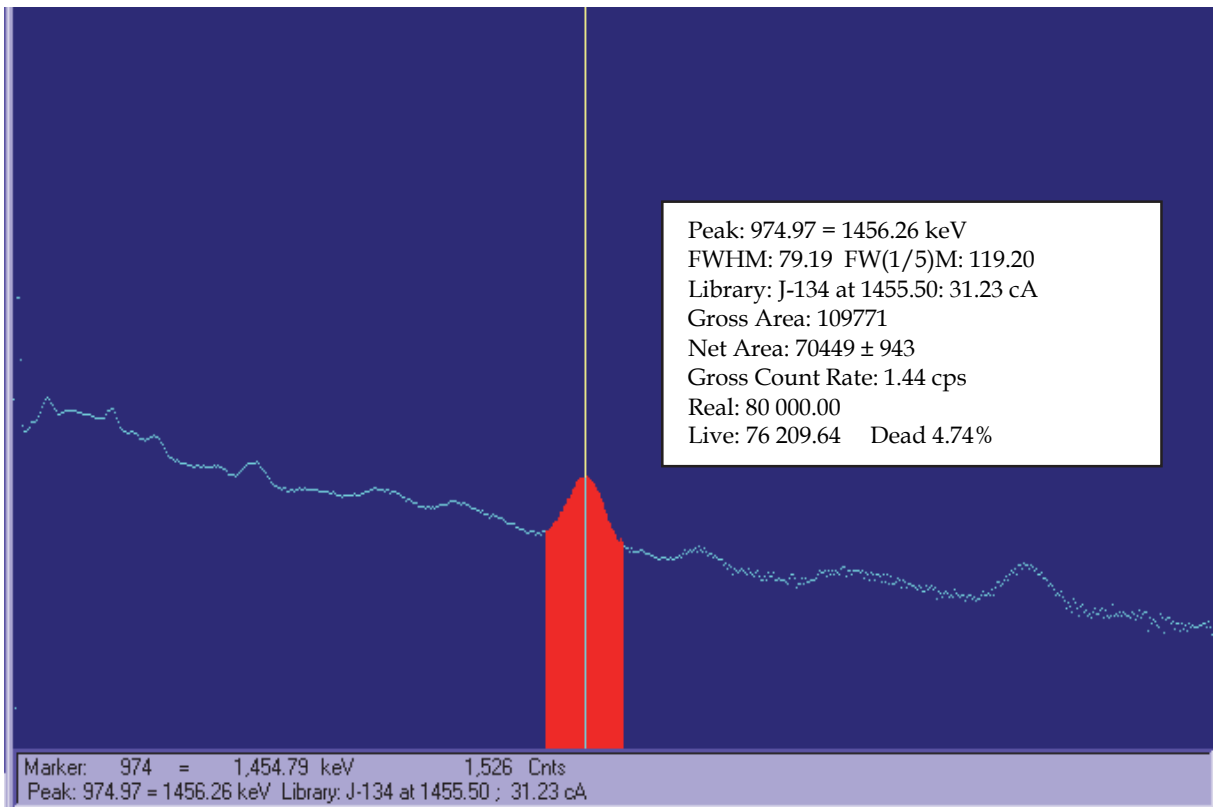


Fig. 9. Gulf of Mexico North East, sea salt spectrum sample, Scintillation Detection System

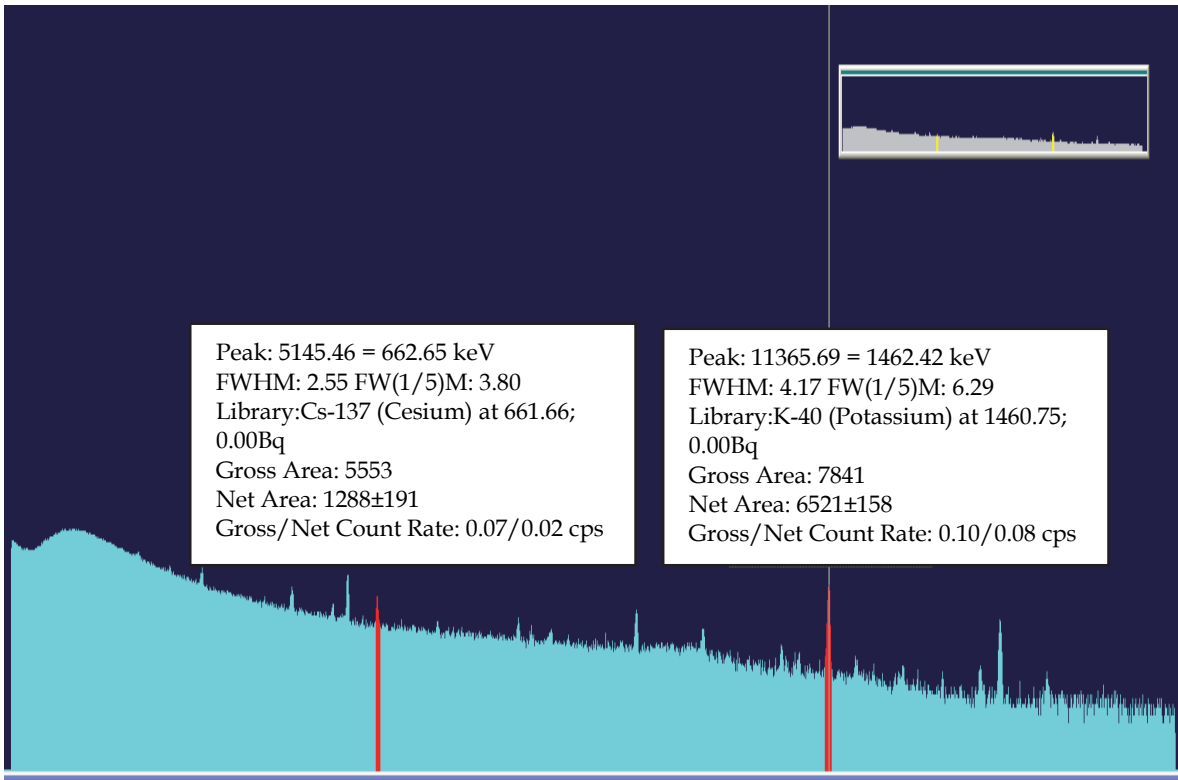


Fig. 10. Background spectrum in HPGe Detection System

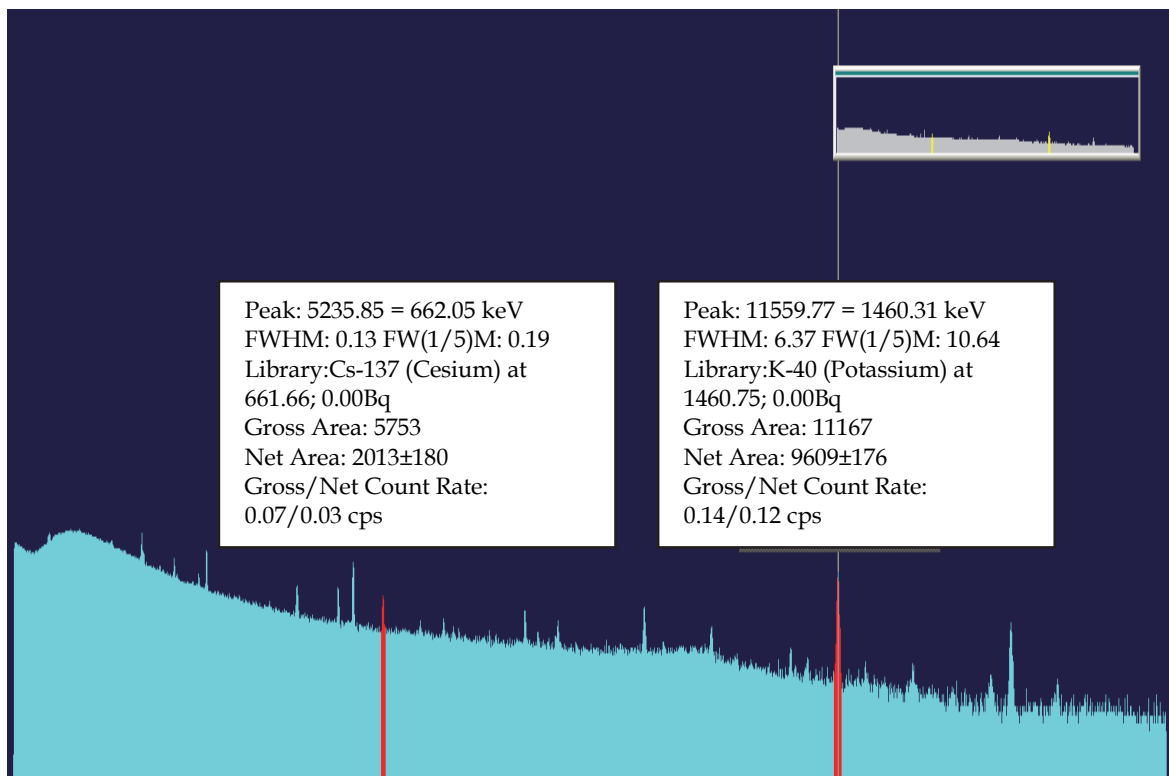


Fig. 11. Gulf of Mexico North East sediment spectrum sample, HPGe Detection System

Sea Salt Samples					Marine Sediment Samples
	Bq ⁴⁰ K/g salt	Bq ⁴⁰ K/L sea water	g salt/L sea water	%K in sea salt	$\%R_{CF} = \frac{Bq^{137}Cs/g}{Bq^{40}K/g} \times 100$
Gulf of Mexico South East	0.276	10.1	36.7	0.88	0.89
Pacific Ocean North	0.073	2.5	34.8	0.23	0.58
Gulf of Mexico North East	0.173	7.3	42.5	0.55	0.93

Table 7. Results of natural radioactivity (⁴⁰K) in sea salt samples and %RCF in marine sediment samples

5. Conclusion

Conclusion of research results is based in several points, however reduced in samples number and extent too, when referring to very large littorals at Mexico.

- a. It seems that radioactive pollution started on the planet at 1945, when first world war was finishing, with the first test of nuclear explosion in Alamo Gordo, followed by war actions in Hiroshima and Nagasaki, and few years later a second test in Bikini atoll.
- b. Since then, a certain number of the so called industrialised countries have performed several tests in different regions of earth, including underground and submarine nuclear explosions.
- c. Also, some accidents in research and power nuclear installations have taken place, notably those in Three Mile Island, USA, Chernobyl, Russia, and lately Fukushima, Japan.
- d. Due to the fact that sea occupies about 80% of planet surface, every pollutant event has a larger probability to reach the sea than any other continental or insular region, starting from the point it has happened.
- e. As growing demand of energy started in societies all over the world in XVIII century, when vapour machine was invented, and today nuclear energy seems to be the most powerful and suitable option to fill up energy demand, closely related to economical development, it looks like already existing, man created radioactive background, presents a strong tendency to grow up in future, since we can not neglect the possibility of accidents as such mentioned before, and even deliberate nuclear explosions as war actions.
- f. It is proposed then, a method to size up the importance and growing rate of radioactive pollution all over the world, by comparing the artificial radioactivity of fission product ^{137}Cs , with that of natural radioisotope ^{40}K , both present in marine sediments at 60-80 meters depth on a great portion of sea bottom.
- g. This procedure seems to be much more general than that to detect just ^{137}Cs in some vegetables such as lichens, which concentrate selectively elementary Cs, and it might be a suitable complement to it.
- h. In this context, already existing radioactive pollution, seems quite possible to detect as a background in marine sediments, since ^{137}Cs half life is 30.07 years, and so it has decayed a little more than 2 half lives, about one fourth of the initial polluting radioactivity disseminated in 1945, plus the following nuclear tests and accidents.
- i. Even when mathematical studies about dispersal of polluting radioisotopes have been successfully applied for limited conditions at a very small fraction of the huge sea (Periañez a, 2004), (Periañez b, 2004), (Periañez c, 2010), it seems that this matter must be verified and treated in a quite empirical way, since natural and polluting radioactivity are facts concerning the whole planet.
- j. In our samples appeared also some other peaks, such as that corresponding to ^{208}Tl (2614 Kev), with very poor resolution in the scintillation counter. Nevertheless, it is indicating the presence of other natural radioisotopes, because it is the last link of the ^{232}Th radioactive chain, in secular equilibrium with its parent and about 11 ancestors decaying at the same rate, before its own decaying to stable ^{208}Pb , with half life of just 3.1 minutes. Then, as a previous link in the chain, it is ^{228}Ac , γ rays emitter with 1459 Kev, and in consequence with possible contribution to ^{40}K peak (energy 1461 Kev) (Lavi, 2004). But as the difference of activity between these two peaks results so large in our samples ($^{40}\text{K}/^{208}\text{Tl} > 10$), then the possible contribution of ^{228}Ac peak (1% branching ratio) to that of ^{40}K (11% branching ratio) results negligible compared with our rather large calculated statistical variation.

- k. Then, and based on previous points, we can say that every large sea portion might be suitably characterized by the percentage of K present in their salts. This can be made very easily in any sea of the planet, by picking up samples from the water surface near the coast. If polluting ^{137}Cs radioactivity (γ rays 662 Kev) is found out accompanying natural radioactivity from ^{40}K (γ rays 1461 Kev), the symptom is present of a rather recent polluting event, whose importance or extent might be evaluated at once by means of the ratio of specific activity per gram of sea salt, or litre of water, of polluting, divided by natural radioactivity and multiplying by 100 in order to have a percentage ($\text{Bq}^{137}\text{Cs} \times 100 / \text{Bq}^{40}\text{K}$). This figure should be concernedly in the measure it approaches to 100%, which means same polluting radioactivity than natural one, and probably it might be useful to avoid the social panic. While same calculation applied to marine sediments 60-80 metres depth, should be useful to measure the already existing background polluting radioactivity, the rate of growing and the real possibility to keep it between tolerable limits.

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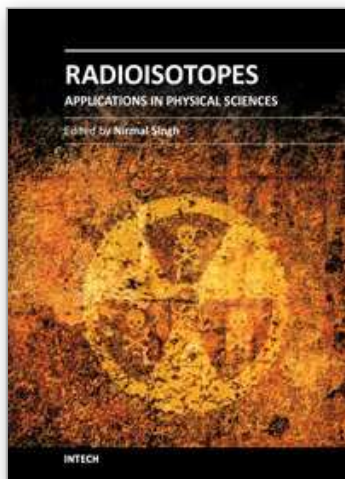
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The book Radioisotopes - Applications in Physical Sciences is divided into three sections namely: Radioisotopes and Some Physical Aspects, Radioisotopes in Environment and Radioisotopes in Power System Space Applications. Section I contains nine chapters on radioisotopes and production and their various applications in some physical and chemical processes. In Section II, ten chapters on the applications of radioisotopes in environment have been added. The interesting articles related to soil, water, environmental dosimetry/tracer and composition analyzer etc. are worth reading. Section III has three chapters on the use of radioisotopes in power systems which generate electrical power by converting heat released from the nuclear decay of radioactive isotopes. The system has to be flown in space for space exploration and radioisotopes can be a good alternative for heat-to-electrical energy conversion. The reader will very much benefit from the chapters presented in this section.

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