

## Project Report

# Distribution of Natural Radioactivity, $^{137}\text{Cs}$ , $^{90}\text{Sr}$ , and Plutonium Isotopes in a Water Column and Sediment Core along the Algerian Coast

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Received 12 March 2007; Accepted 27 November 2007

Recommended by Luigi Petrizzi

Concentrations of natural ( $^{40}\text{K}$ ,  $^{210}\text{Pb}$ , uranium, and thorium series) and artificial radioelements ( $^{137}\text{Cs}$ ,  $^{90}\text{Sr}$ ,  $^{239+240}\text{Pu}$ ) were determined in seawater and sediment samples collected from stations along the Algerian coast. Seawater was collected from the surface to a maximum depth of 2000 m; the sediment cores were sampled from a depth of around 1500 m. This work was carried out in August 2001, in the framework of the Regional African project /7/004 (RAF), by the accomplishment of an oceanographic campaign organized by the International Atomic Energy Agency (IAEA) in collaboration with Commissariat à l'Énergie Atomique (COMENA) and L'Institut des Sciences de la Mer et de L'Aménagement du Littoral (ISMAL), on board of the research vessel of M.S. Benyahia of ISMAL. In addition to the record of the conductivity (mS) and temperature ( $^{\circ}\text{C}$ ) data at each station, seawater samples were treated and preconcentrated on board, those of sediment cores were divided into different layers in order to undergo analyses in the laboratory of radiological impact studies of Algiers. Concentration results were obtained for  $^{137}\text{Cs}$  and  $^{90}\text{Sr}$  in mBq/L and  $^{239+240}\text{Pu}$  in  $\mu\text{Bq/L}$  in seawater, and also for natural and artificial radionuclides in Bq/kg dry weight in the layers of the sediment cores. The different profiles of  $^{137}\text{Cs}$ ,  $^{239+240}\text{Pu}$ ,  $^{90}\text{Sr}$ , and  $^{210}\text{Pb}$  concentrations against depth were presented to show artificial radioactivity distribution in the water column and sediment core. Concentrations of  $^{137}\text{Cs}$  in the sediment core were also used to identify the  $^{137}\text{Cs}$  peak in the area of interest.

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## 1. INTRODUCTION

Natural radioactivity is present since the creation of earth, that is, billions of years, and is introduced into the marine environment through terrestrial and atmospheric routes. The most important radioelements are  $^{40}\text{K}$  and uranium and thorium series known to have a very long half-life and to be abundant in the environment. In addition to that, and since the introduction of the nuclear technology, several artificial radioactive sources were added to the marine radioactive inventory. These sources result from nuclear weapons tests, authorized releases from nuclear facilities, and also from nuclear accidents, particularly that of Chernobyl occurred in April 1986. According to the physicochemical properties of the receiving medium and those of the radioelements of interest, radioactivity can be introduced through several routes into the marine environment. Radioactivity can be both directly introduced into the marine environment by wet or dry depositions or from the terrestrial medium, and dispersed by different processes depending on physicochemical properties of the medium and the radioelements of in-

terest. Radioactivity can be diluted, transported, fixed, distributed, and deposited on the marine bottom, and much information can be deduced to determine the behavior and advance of radioelements in the marine environment. Therefore, it is necessary to measure the concentration of some radioelements of interest, namely,  $^{137}\text{Cs}$ , Pu isotopes,  $^{90}\text{Sr}$ , and  $^{210}\text{Pb}$ , in some representative samples such as seawater and sediment. In this work, and in the framework of the radiological surveillance along the Algerian coast and the study of marine process, several oceanographic campaigns were organized. Concentrations of  $^{137}\text{Cs}$  reported in 1997 and 1999 along the Algerian coast show concentrations ranging from  $2.05 \pm 0.09$  to  $3.15 \pm 0.34$  mBq/L with an average value of  $2.59 \pm 0.26$  mBq/L in seawater, and from  $0.1 \pm 0.04$  to  $6.29 \pm 0.3$  Bq/kg in sediment for  $^{239+240}\text{Pu}$ ; the concentrations range from  $0.16 \pm 0.02$  to  $1.02 \pm 0.09$  Bq/kg [1]. Concerning natural radioactivity, concentrations of  $^{40}\text{K}$  range from  $312 \pm 14.6$  to  $613 \pm 25$  Bq/kg. Regarding the results of the sampling campaign of August 2001 of the present work,  $^{137}\text{Cs}$ ,  $^{90}\text{Sr}$ , Pu isotopes, and  $^{210}\text{Pb}$  concentrations in Bq/kg were determined in several sediment cores and seawater columns,

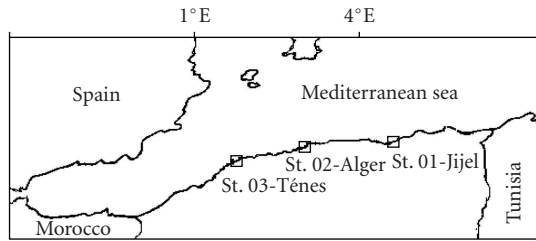


FIGURE 1: Sampling locations.

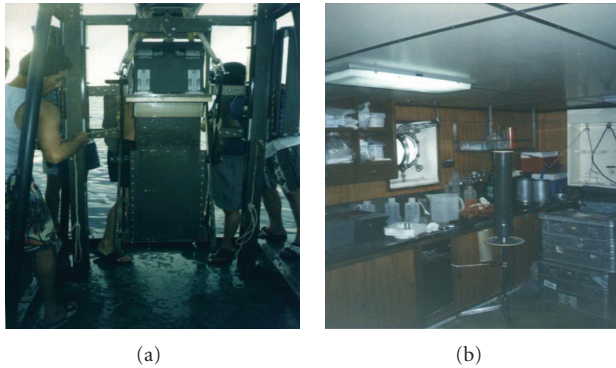


FIGURE 2: (a) A box corer for the collection of sediment, and (b) a device for sectioning sediment, on board of the research vessel of M.S. Benyahia-2001.

and their profiles versus depth (m) were also plotted. The average concentrations of  $^{137}\text{Cs}$  and  $^{90}\text{Sr}$ , for example, in surface seawater for the year 2000 in the same area, are about 2.5 mBq/L [2–4] and about 1.5 mBq/L [5], respectively.

## 2. MATERIALS AND METHODS

### 2.1. Sampling

In the framework of the radiological surveillance program of the marine environment along the Algerian coast and the IAEA regional project RAF/7/004, an oceanographic campaign was organized by the IAEA in collaboration with COMENA and ISMAL, in August 2001, on board of the research vessel M.S. Benyahia of ISMAL, in order to collect sediment cores and seawater columns at different stations at the center, the east, and the west (Figure 1) and also to record some marine data such as temperature ( $^{\circ}\text{C}$ ) and conductivity (mS). The purpose of this campaign is to measure  $^{137}\text{Cs}$ ,  $^{90}\text{Sr}$ , and Pu isotopes concentrations in a seawater column and that of  $^{137}\text{Cs}$  and  $^{210}\text{Pb}$  in the different layers of the sediment core.

On board of M.S. Benyahia research vessel, seawater samples were collected at different depths using a stainless steel bottle of a volume of 250 L, where an amount of 150 L was filtered on a  $0.45\ \mu\text{m}$  membrane filter and transferred to a tank of polyethylene, in order to precon-

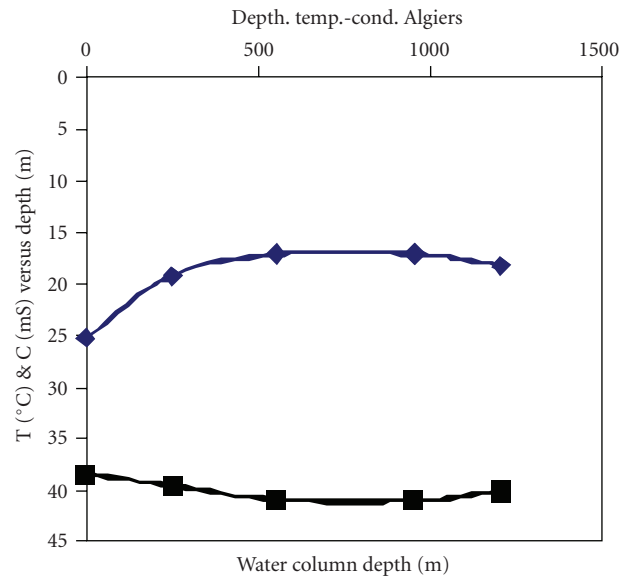
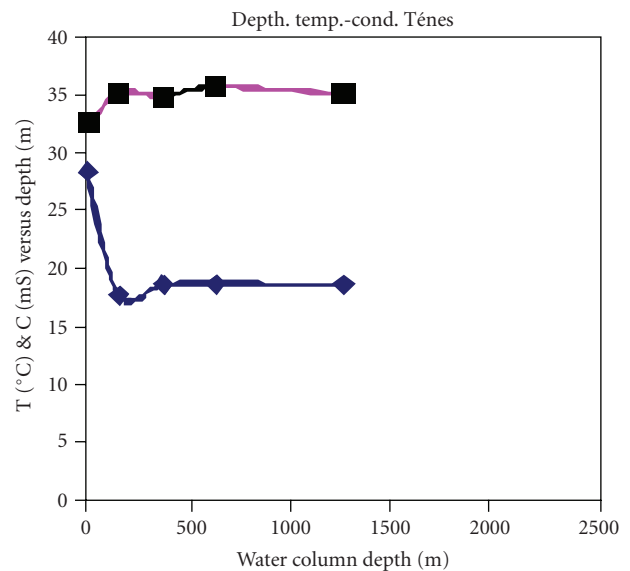
(a) Profile of T ( $^{\circ}\text{C}$ ) and C (mS) versus depth (m) (station 02, Algiers, 2001)(b) Profile of T ( $^{\circ}\text{C}$ ) and C (mS) versus depth (m) (station 03, Ténes, 2001)

FIGURE 3

centrate the radioelements of interest, namely,  $^{137}\text{Cs}$ ,  $^{90}\text{Sr}$ , and Pu isotopes, for a radiochemical analysis in the laboratory.

Concerning sediment samples, an IAEA box corer (see Figure 2(a)) was used to collect sediment cores at depths reaching a maximum of 1500 m. Sediment cores were carefully sectioned into slices on board with an appropriate device to cut into layers of 0.5, 1, and 2 cm of thickness; see Figure 2(b). Sediment samples were stored in a freezer to determine porosity and to count them by gamma spectrometry.

TABLE 1: Measurement of T (°C) and C (mS) parameters at stations 02 and 03, Algiers and Ténés sampling campaign. (08 to 23 August 2001.)

Station 02, Algiers/depth (m)	T (°C)	C (mS)	Station 03, Ténés/depth (m)	T (°C)	C (mS)
0	25.3	38.4	0	28.3	32.4
250	19.2	39.6	250	17.7	35.2
550	17.1	40.9	600	18.6	34.9
950	17.1	41	1000	18.7	35.7
1200	18.2	40.1	2000	18.6	35.1

TABLE 2: Concentration of  $^{137}\text{Cs}$  along water column in Ténés and Algiers stations.

Concentration (mBq/L)	$2.78 \pm 0.12$	$2.1 \pm 0.13$	$1.8 \pm 0.11$	$2.2 \pm 0.13$	$1.31 \pm 0.08$
Ténés station/depth (m)	0	250	600	1000	2000
Concentration (mBq/L)	$2.25 \pm 0.15$	—	$2.31 \pm 0.15$	$2.21 \pm 0.14$	$2.33 \pm 0.15$
Algiers station/depth (m)	—	—	550	950	1200

Porosity is defined as the part of the water content in the whole sediment of analysis [6].

## 2.2. Radioactivity measurement

Seawater samples under ammonium molybdophosphate precipitate (AMP) were put in plastic beakers of 100 cc volume to be analyzed by direct counting by gamma spectrometry using a high-purity germanium detector of 23% relative efficiency and 1.8 keV at 1332 keV energy peak of  $^{60}\text{Co}$ . Samples were put in contact with the detector and counted for 24 hours to 72 hours in order to reach reliable statistic counting. Detection efficiency was determined by preparing a standard sample in the same density and geometry conditions by introducing a liquid source of  $^{134}\text{Cs}$ . The main contribution to the efficiency uncertainty is the standard solution error added to the statistic error.

With regard to sediment samples, they were dried at 80°C in the oven, crushed into fine powder, homogenized, and transferred to cylindrical plastic beakers of 100 cc volume. They were counted and analyzed with the same HP germanium detector, using a Genie-2000 and an interwinner program to process energy spectra. Detection efficiency was also determined using the same geometry conditions by contamination of the standard using a radioactive liquid source of  $^{152}\text{Eu}$  to cover the range of energy of interest. The main contribution to the concentration uncertainty is the efficiency error plus the statistic error.

Natural and artificial gamma emitting radioelements ( $^{210}\text{Pb}$ ,  $^{137}\text{Cs}$ ) were conditioned and evaluated by gamma spectrometry.

However, for  $^{90}\text{Sr}$  and  $^{239+240}\text{Pu}$ , samples undergone radiochemical separations,  $^{90}\text{Sr}$  was extracted using selective crown ether resin, and concentration was determined by liquid scintillation counting through a source prepared by gravimetric precipitation using a stable carrier of  $^{90}\text{Sr}$  as strontium nitrate. For  $^{239+240}\text{Pu}$ , the extraction was carried

out by anion exchange, and the source was prepared by coprecipitation with neodymium fluoride, using a radioactive tracer ( $^{242}\text{Pu}$ ), and evaluated by alpha spectrometry.

## 3. RESULTS AND DISCUSSION

Sampling locations of seawater column and sediment cores are presented in Figure 1. Records data of temperature (°C) and conductivity (mS) are given in Table 1 and their profiles versus depth are shown in Figures 3(a) and 3(b). Concentration levels of  $^{137}\text{Cs}$  in mBq/L in the water column of Algiers and Ténés stations are indicated in Table 2, where uncertainty used is  $1\sigma$ .  $^{137}\text{Cs}$  concentration in mBq/L at the surface water and at different depths of Ténés station ranges between  $1.31 \pm 0.08$  and  $2.20 \pm 0.13$  mBq/L, with a peak at 1000m, and that of Algiers station ranges from  $2.21 \pm 0.14$  to  $2.3 \pm 0.15$  mBq/L. Regarding  $^{90}\text{Sr}$  and  $^{239+240}\text{Pu}$  in the station of Ténés, the average concentration in seawater is  $4.79 \pm 0.5$  mBq/L and  $18.01 \pm 2.58$   $\mu\text{Bq/L}$ , respectively. The obtained results of  $^{90}\text{Sr}$  and  $^{239+240}\text{Pu}$  in the station of Ténés are given in Tables 3 and 4.  $^{137}\text{Cs}$  profiles versus depth in both stations are plotted and shown in Figures 4(a) and 4(b). The average values of  $^{137}\text{Cs}$  concentration in Algiers and Ténés stations are  $2.28 \pm 0.15$  mBq/L and  $1.89 \pm 0.11$  mBq/L, respectively. These values were compared to other works (values ranging from 2.04 mBq/L to 4.4 mBq/L) in the same area, and found to be in the same range [2–5, 7].

Also  $^{90}\text{Sr}$  and  $^{239+240}\text{Pu}$  profiles were plotted against depth for Ténés station, and shown in Figures 5(a) and 5(b).  $^{239+240}\text{Pu}$  concentrations are in the range of those given by different authors; however, those of  $^{90}\text{Sr}$  seem to be overestimated. This could be explained by some systematic errors that might have been undertaken during the radiochemical analysis and not due to a contamination.

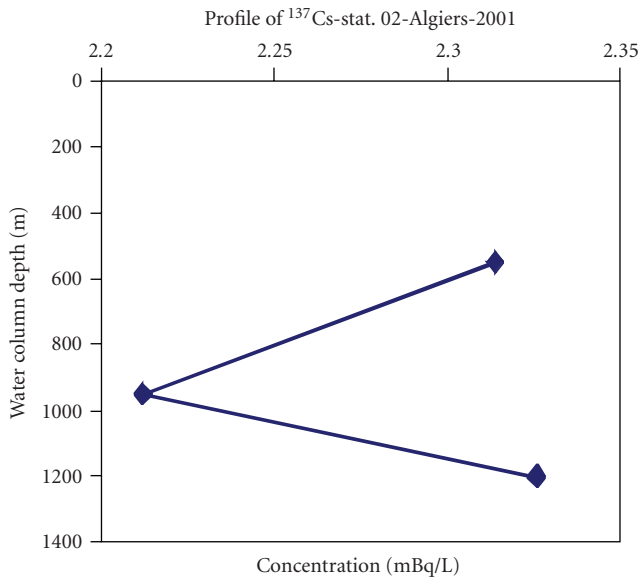
Concerning sediment cores, concentrations of  $^{137}\text{Cs}$  vary from 1.26 Bq/kg dry weight to 11.69 Bq/kg dry weight in Jijel station. Concentrations in Bq/kg versus depth of the different sediment core layers gave  $^{137}\text{Cs}$  profiles in Algiers and Jijel

TABLE 3: Concentration of <sup>90</sup>Sr along seawater column of Ténés station, 2001.

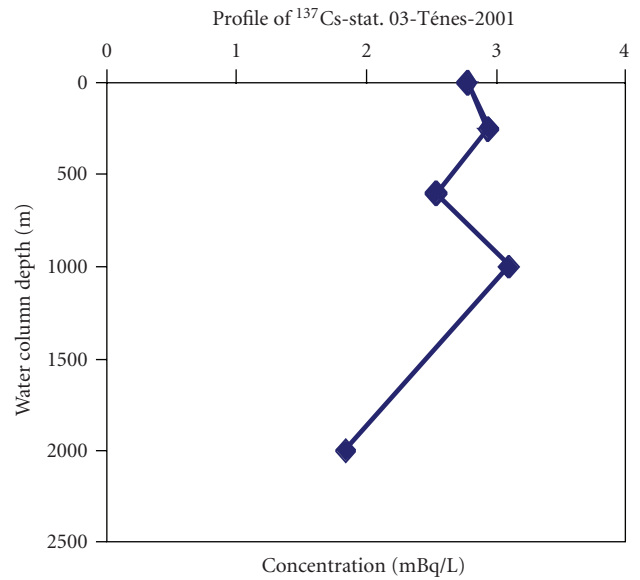
Concentration (mBq/L)	1.75 ± 0.2	9.6 ± 1.0	3.45 ± 0.4	5.5 ± 0.6	3.65 ± 0.5
Ténés station/depth (m)	0	250	600	1000	2000

TABLE 4: Concentration of <sup>239+240</sup>Pu along seawater column of Ténés station, 2001.

Concentration (μBq/L)	8.2 ± 1.0	18.5 ± 2.7	16.00 ± 2.4	25.5 ± 3.7	22.0 ± 3.1
Ténés station/depth (m)	0	250	600	1000	2000

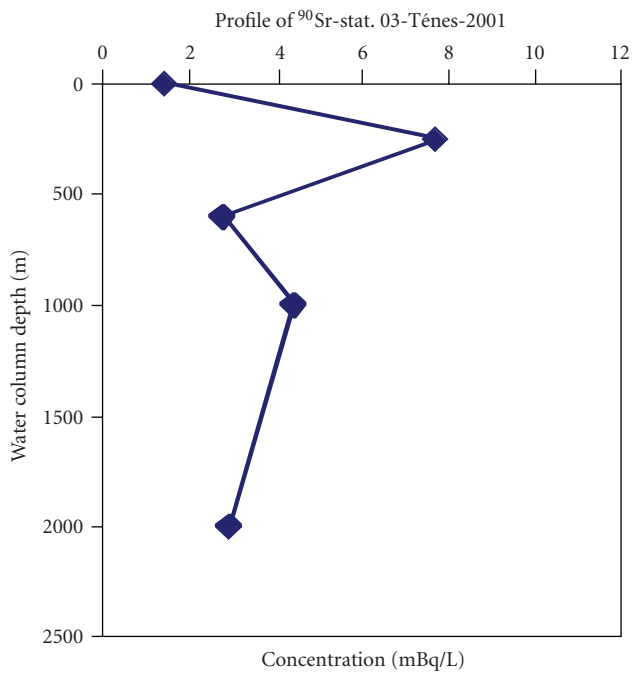


(a) Profile of <sup>137</sup>Cs versus depth (m) (station 02, Algiers, 2001)

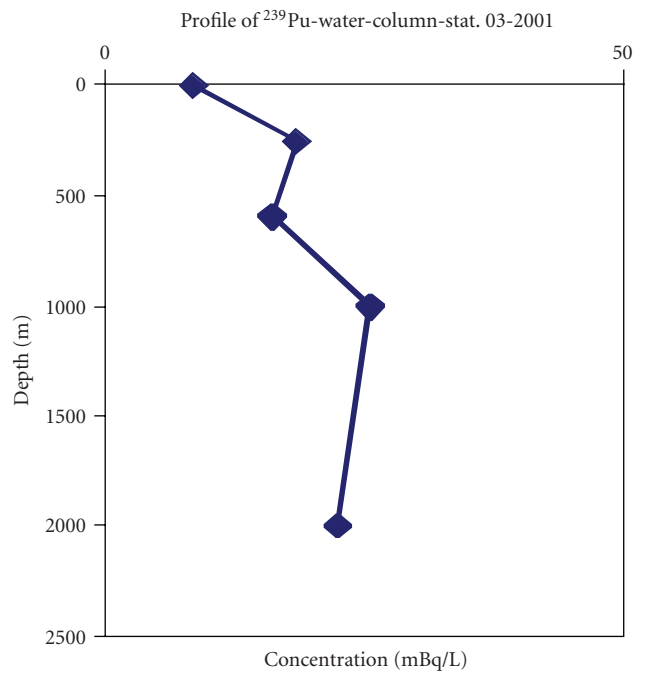


(b) Profile of <sup>137</sup>Cs versus depth (m) (station 03, Ténés, 2001)

FIGURE 4



(a) Profile of <sup>90</sup>Sr versus depth (m) (station 03, Ténés, 2001)



(b) Profile of <sup>239+240</sup>Pu versus depth (m) (station 03, Ténés, 2001)

FIGURE 5

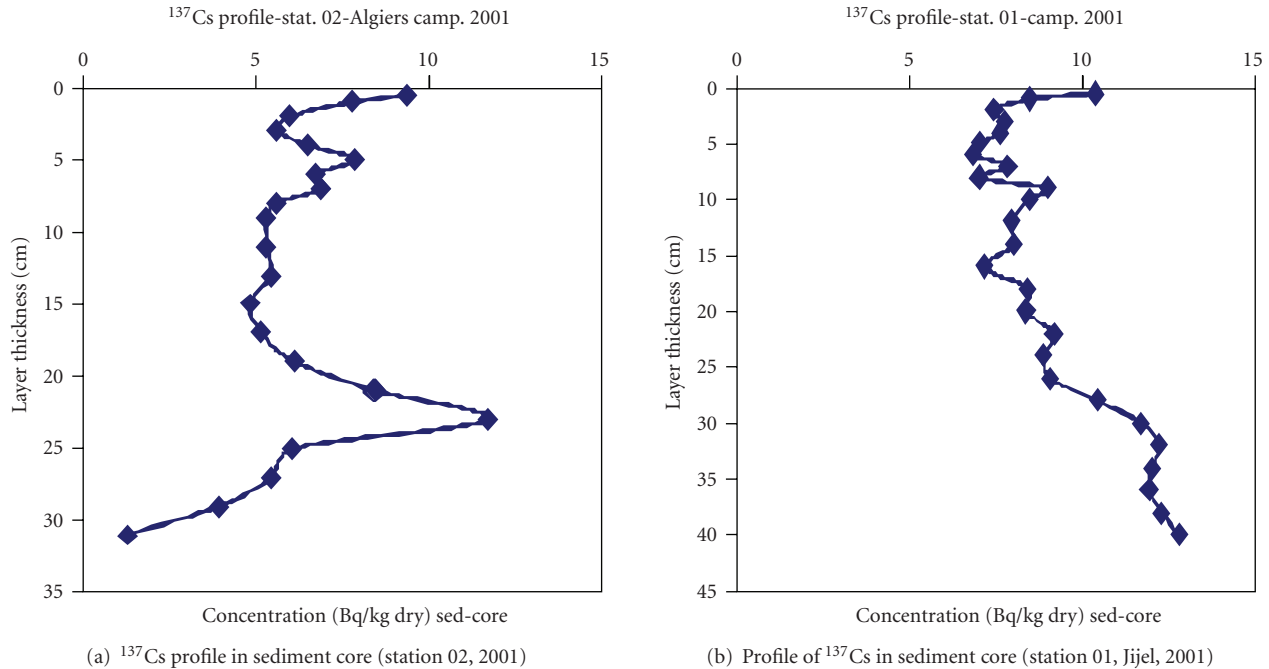


FIGURE 6

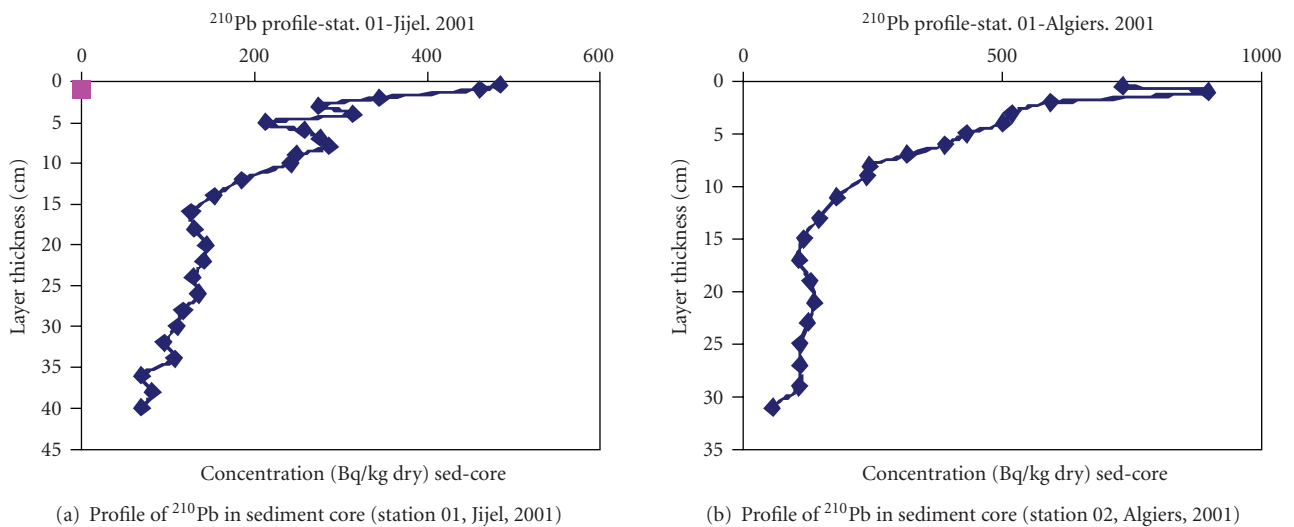


FIGURE 7

stations, as shown in Figures 6(a) and 6(b). In the same way,  $^{210}\text{Pb}$  concentrations were determined in Algiers and Jijel stations, with values ranging from 57 to 895 Bq/kg and from 69 to 484 Bq/kg dry weight, respectively. Concentrations in Bq/kg dry weight were also plotted against depth of sediment core layers to show  $^{210}\text{Pb}$  profile in Algiers and Jijel stations as indicated in Figures 7(a) and 7(b).

#### 4. CONCLUSIONS

This study enabled us to determine natural and artificial radioelements concentration.  $^{137}\text{Cs}$ ,  $^{90}\text{Sr}$ , and  $^{239+240}\text{Pu}$  distribution is observed in a water column showing subsurface

peaks at different layers (250 m and 1000 m depth). These peaks might belong to 1963 nuclear tests and Chernobyl accident. These results confirm the behavior of  $^{137}\text{Cs}$  as conservative and Pu isotopes adsorbed by suspended particles.

Concerning  $^{137}\text{Cs}$  and  $^{210}\text{Pb}$  concentrations in the sediment core, they could be used to estimate the radioactive inventory and the sedimentation rate in the area of study.

#### ACKNOWLEDGMENTS

The authors would like to thank the International Atomic Energy Agency for the organization of the sampling campaign in the framework of the RAF/7/004, L'Institut des

Sciences de la Mer et de l'Aménagement du Littoral (ISMAL) for placing Mohamed Seddik Benyahia at our service, and the assistance of our colleagues participating in the cruise during seawater sampling.

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