

# Evaluation of the sedimentation rate with $^{210}\text{Pb}$ and $^{137}\text{Cs}$ using the CRS model and estimation of the total inventory of sediment in the Algerian Coast

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## Abstract

Lead-210 and to a lesser extent  $^{137}\text{Cs}$  are the most widely used radionuclides in soil erosion and recent radiogeochronology studies. This is basically due to their appropriate half-life, suitable for studying sedimentary processes occurring in the last 100 – 150 years. In coastal systems, sediments are transported by rivers to the coastal ocean, deposited under the influence of several marine processes. A study was carried along the Algerian coast, between 1999 and 2004, under the framework of the IAEA regional project RAF/7/004. Sediment cores were collected from different stations as given in figure 1 on board of the research vessel of M.S. Benyahia of ISMAL in collaboration with COMENA and IAEA, in order to study sediment accumulation in this area.

For this purpose,  $^{210}\text{Pb}$  and  $^{137}\text{Cs}$  were determined in sediment sections, using direct counting by gamma spectrometry analysis. The accuracy of their quantification is mainly dependent on the errors including ambient background variation, attenuation of the  $^{210}\text{Pb}$  45 keV gamma ray in the sample and efficiency calibration. Vertical distribution of  $^{210}\text{Pb}$  and  $^{137}\text{Cs}$  in sediments cores were used as a tool to establish the sediment chronology and to estimate the total inventory in the coastal environment. Concentrations in Bq/kg dry weight of  $^{210}\text{Pb}$  and  $^{137}\text{Cs}$  were determined in the four sediment cores. For concentration levels of  $^{137}\text{Cs}$  it ranges from  $(1.3\pm 0.1)$  Bq/Kg to  $(12.8\pm 0.8)$  Bq/ Kg with an average value of 7.5 Bq/Kg, and for  $^{210}\text{Pb}$  concentrations in Bq/Kg dry weight it was found to be in the range of  $(57\pm 5$  to  $895\pm 98)$  Bq/Kg dry weight. The estimated sedimentation rates deduced from the  $^{210}\text{Pb}$  and  $^{137}\text{Cs}$  concentrations profiles (some of them are presented in figure 2) and using the CRS model [1], range from 20 mm/yr to 27 mm/yr. Sedimentation rates and total inventories were determined at each station and maximum peaks were identified. Data obtained in this work were compared to those carried out in the same area by other authors, [2], [3].

Keywords: *Sedimentation rate, CRS model,  $^{210}\text{Pb}$ ,  $^{137}\text{Cs}$ , Algerian Coast, Gamma Spectrometry*

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## 1. Introduction

The main contamination sources are those resulting from nuclear fallout of nuclear tests carried out since 1945 with a peak between 1952-1963, releases from nuclear installations and nuclear

accidents. The most important nuclear accident is that of Chernobyl occurred in April 1986, with an important release of radioactive materials to the atmosphere, especially  $^{137}\text{Cs}$ . Evaluation of this contamination becomes necessary through development and use of different techniques in order to quantify it using representative samples.

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Algerian coasts in particular, are under the influence of water exchanges from the Atlantic Ocean through the strait of Gibraltar, water coming from other regions of the Mediterranean Sea. This area was not studied to evaluate the radioactive contamination or study its distribution in the marine medium.

This sea is well known for heavy maritime traffic with possible accidents including nuclear submarines and it has been recommended by the International Atomic Energy Agency (IAEA) in collaboration with COMENA and ISMAL to carry out a regional technical cooperation project with the aim of assessing the present radionuclide levels in the region along the Algerian coast. The results could be used as background values for radiological assessments in the case of emergency situations, but also for marine environmental studies using radionuclides as traces of movement of water masses and of biogeochemical processes in the water column and in sediments (e.g. Livingston and Povinec, 2000).

Except for natural radionuclides of U and Th and their decay products, anthropogenic radionuclides representing global fallout from atmospheric nuclear tests, carried out mostly from the late 1950s until the early 1960s, have also been frequently used in marine studies (e.g. Livingston and Povinec, 2002). Several radionuclides (e.g. 3H, 14C, 90Sr, 137Cs, Pu isotopes) were produced in large quantities with a distinct peak observed in the atmosphere in 1963 and in seawater in 1964 (e.g. Povinec et al., 2005), and contributed to contaminate the eastern and northern Mediterranean basin (UNEP/IAEA, 1992). A better understanding of marine processes, water movement and pollutants behaviour, is however, important for a sustainable development of coastal area and their protection against any radioactive or non-radioactive contamination.

For the achievement of this study in order to evaluate the radioactive level and distribution of radioactivity along the Algerian coast, we proceeded in collaboration with ISMAL and IAEA, to the organization of several oceanographic campaigns between 1997 and 2004. Samples analysed and presented in this work are sediment cores collected

from different depths from the seawater surface.

The objectives presented in this part of work are:

- Determination of radioactive reference level (mainly 137Cs) in surface sediment, to use it as a basic data in case of any accidental event;
- Determination of 137Cs and 210Pb activity in a sediment core to study their distribution from vertical profiles;
- Estimation of sedimentation rate using 137Cs and 210Pb massic activity;
- Estimation of total inventory (sediment core) of natural and artificial radioactivity along the Algerian coast.

## 2. EXPERIMENTAL METHODS

### 2.1 Sampling and conditioning of samples

Four Oceanographic campaigns were carried out in the south-western Mediterranean sea along the Algerian coasts. The first ones were organised by COMENA and ISMAL in 1997 and 1999, and the two others (in 2001 and 2004) by the IAEA in collaboration with COMENA and ISMAL, all on board of the research vessel boat of "Mohamed Seddik BENYAHIA" of ISMAL. Sampling locations of the four oceanographic campaigns are those presented in Figure 1.

Sediments were collected using different corers during the four oceanographic campaigns. Core sediments of 50 cm depth were sliced on board into different layers of 0.5 cm, 1cm and 2cm, conditioned and conserved in a freezer for detailed radiochemical analysis in the laboratory. The equipments used are those presented in Figure 2.

### 2.2 Determination of gamma emitting radionuclides concentration.

After conditioning, sediment samples were prepared for direct counting by gamma spectrometry, and detection yield was determined using efficiency calibration curve obtained from preparation of standard sample having the same conditions of volume and geometry, as indicated in Figure 3.

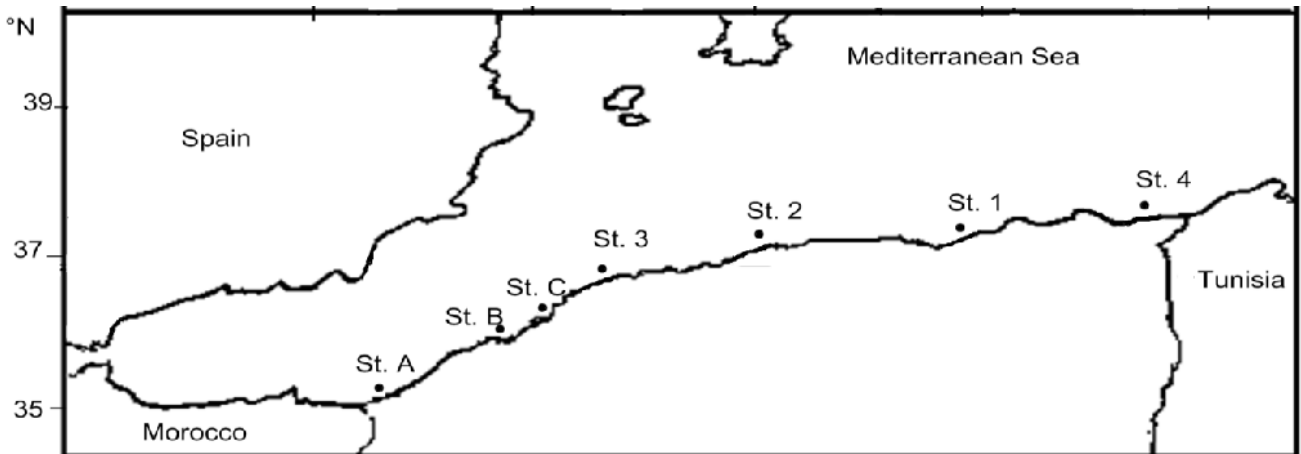


Fig 1- Locations of sediment core samples.

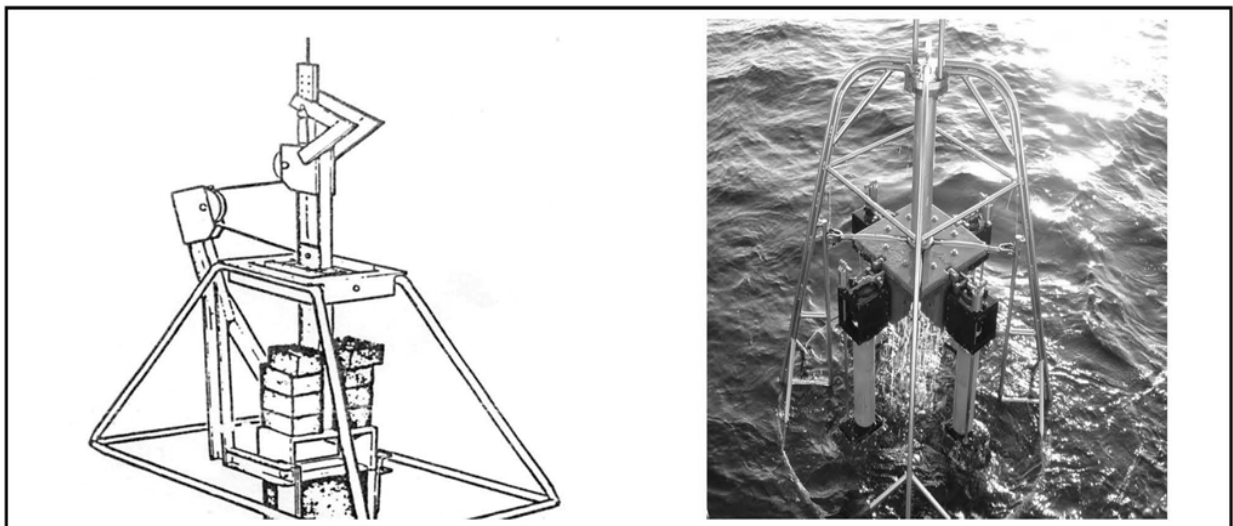


Fig 2- Equipments used for sediment core sampling, in 1999, and 2001, respectively

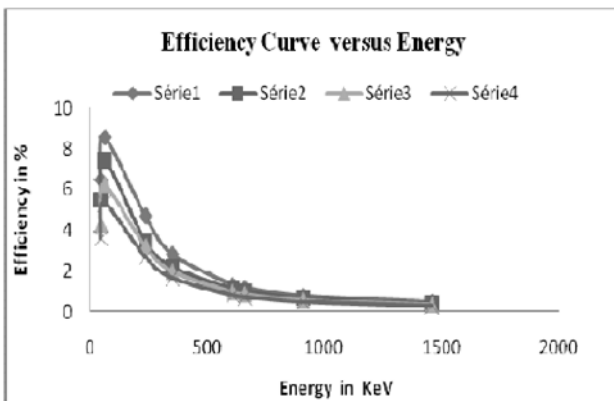


Fig 3- Efficiency curve obtained from preparation on standard sample at the laboratory

Quality of results was assured by participation in IAEA-384 (Fangataufa sediment, Povinec et al., 2007) intercomparison exercises. The total uncertainties of gamma-spectrometry results (statistics, and calibration) were kept below 10%.

### 3. RESULTS

#### *137Cs and 210Pb in sediments*

Specific activities of natural ( $^{210}\text{Pb}$ ,  $^{226}\text{Ra}$ ) and artificial radionuclides ( $^{137}\text{Cs}$ ) in sediment cores collected at stations 1, 2 and 4 at depth of 270 m,

1220 m and 550 m, from the water surface, respectively, are given in figure 4, where vertical distribution of activity was plotted against depth of the core (in cm). Level of massic activity at the surface of sediment core range from  $8.4 \pm 0.7$  to  $9.4 \pm 0.8$  Bq/kg dry weight, with an average value of  $9.0 \pm 0.08$  Bq/kg.

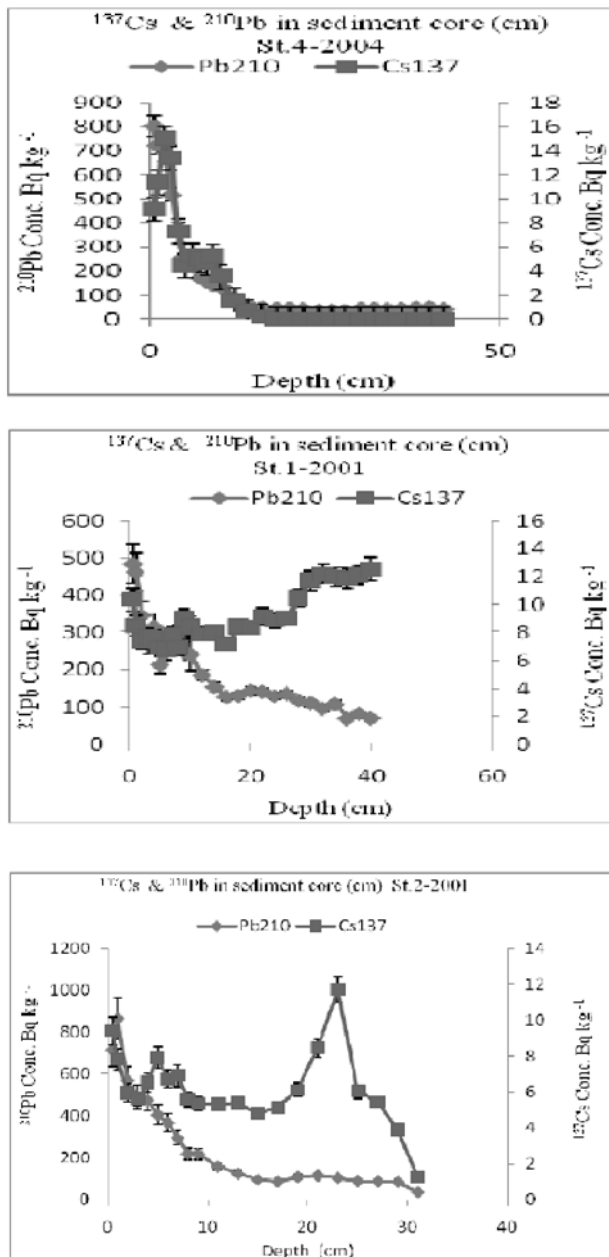


Fig 4- Profiles of  $^{137}\text{Cs}$  and  $^{210}\text{Pb}$  sediment cores at station 1, 2 and 4 collected in 2001 and 2004

Penetration depth of  $^{137}\text{Cs}$  reached easily 40 cm

at station 1, whereas at station 2 and 4, it reached only 30 cm and 15 cm respectively.

The observed  $^{137}\text{Cs}$  peaks are localised at different depths, 4 cm and 21-22 cm at station 2, and 1 cm and 8 cm at station 4. Station 1 showed a very different  $^{137}\text{Cs}$  Profile, having high values at the surface, scattered values at medium depths, and increasing values at the core bottom. Clearly, this  $^{137}\text{Cs}$  profile was significantly affected by bioturbation and mixing processes in the sediment. Naturally occurring  $^{210}\text{Pb}$ , resulting from radioactive decay of  $^{238}\text{U}$  chain, is produced in the atmosphere through  $^{222}\text{Rn}$  decay, and transferred by wet and dry deposition on the earth's surface. The excess  $^{210}\text{Pb}$  in sediment, expressed by  $^{210}\text{Pb}_{\text{ex}} = ^{210}\text{Pb}_{\text{total}} - ^{226}\text{Ra}$  was used to estimate sediment accumulation rate (Cochran et al., 1998; Ueda et al., 2004), on the basis of a constant sedimentation rate.

The excess  $^{210}\text{Pb}$  activity in the sediment at any depth  $x$  of the core is given by

$$A(x) = A_0 e^{-\lambda t}$$

Where  $\lambda$  is the decay constant of  $^{210}\text{Pb}$  and  $A_0$  is the excess  $^{210}\text{Pb}_{\text{ex}}$  activity at the sediment surface. The accumulation rate ( $S$ ) is expressed by

$$S = (\lambda x) / \ln(A_0/A_x)$$

The excess  $^{210}\text{Pb}_{\text{ex}}$  profiles in the sediment core collected from station 1, 2 and 4 were also presented in figure 4. They show similar profile to that of  $^{137}\text{Cs}$ , and we notice that radionuclides of interest reached a relatively important depth in station 1 compared to station 2 and 4.

An exponential radioactive decay of  $^{210}\text{Pb}$  versus depth in the sediment core was also observed at station 4 (Annaba, 2004), giving the lowest sedimentation rate  $0.15 \pm 0.04$  cm  $\text{y}^{-1}$  in the Algerian basin. The observed  $^{137}\text{Cs}$  peaks at 2-3 cm and 8-9 cm (see figure 5) might correspond to Chernobyl and nuclear fallout peaks, respectively.

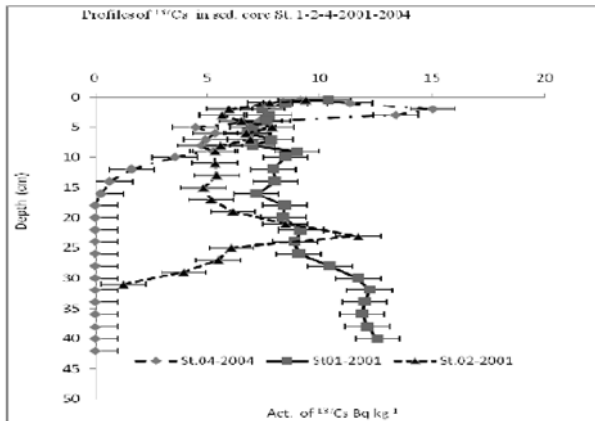


Fig 5- Profiles of <sup>137</sup>Cs in Bq kg<sup>-1</sup> in sediment core versus depth (cm) at stations 1-2- (2001) and 4-(2004).

## 4. DISCUSSION

### 4.1 Sediment

The natural occurring radionuclide <sup>210</sup>Pb is produced in the atmosphere, resulting from the <sup>222</sup>Rn radioactive decay. It is introduced in the marine medium from the soil surface, after dry or wet deposition. The excess <sup>210</sup>Pb is generally used to estimate sediment accumulation rate, assuming a

constant <sup>210</sup>Pb input (Cochran, 1998; Ueda, 2004).

Figure 6 shows <sup>210</sup>Pb and excess <sup>210</sup>Pb profiles in sediment cores of the studied stations, and the reached depth for each station. In some stations, we observe a typical <sup>210</sup>Pb radioactive decay, and in others we observe a perturbation due likely to a bioturbation.

The sedimentation rate was calculated and ranged from 0.15 to 0.7 cm y<sup>-1</sup>. This method is useful and determines chronology of radioactive or non-radioactive contamination in a marine medium.

### 4.2 <sup>137</sup>Cs inventory in sediment

Estimation of <sup>137</sup>Cs inventory in sediments was calculated on the basis of integration of the different parts of the profile of <sup>137</sup>Cs activity distribution versus depth (cm), as indicated in figure 4. Global inventory increase from the western to the eastern part of the Algerian coast, with values of  $180 \pm 20$ ,  $330 \pm 30$ ,  $350 \pm 30$  Bq/m<sup>2</sup> for station 2, 1 et 4, respectively. <sup>137</sup>Cs inventory in sediment follows the same variation as it is in the water column.

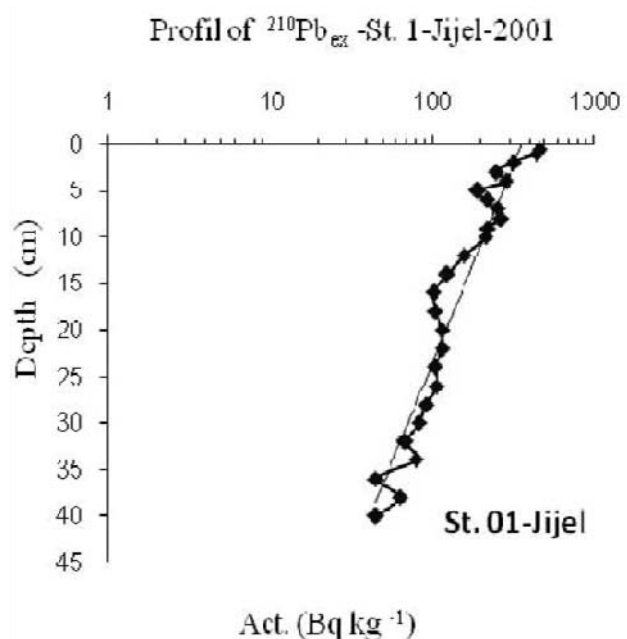
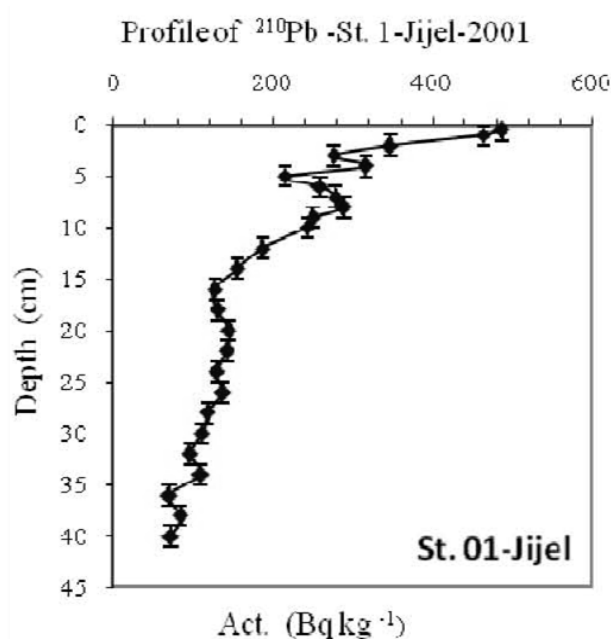


Fig 6- Logarithmic plotting of <sup>210</sup>Pb<sub>ex</sub> versus depth

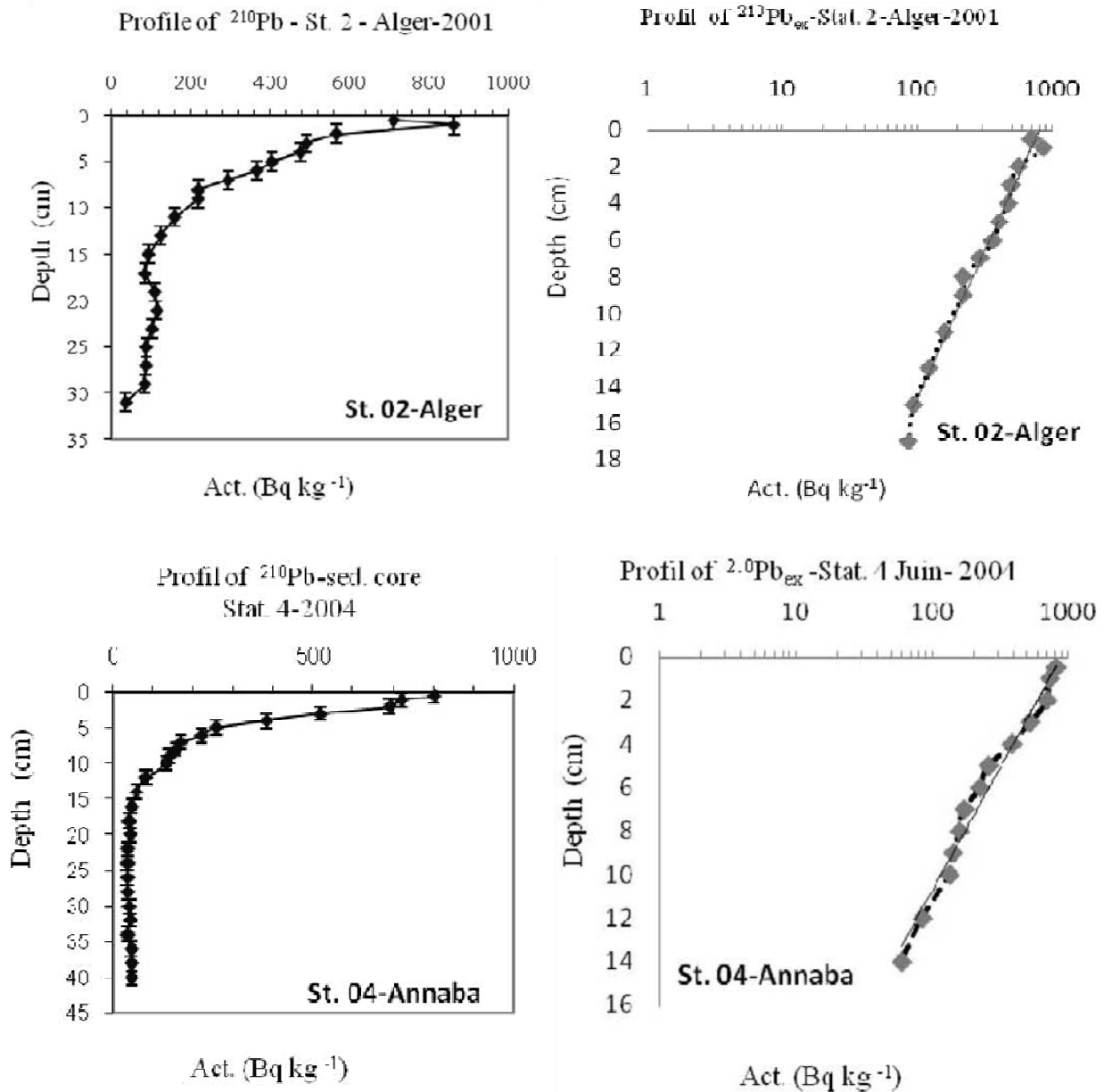


Fig 6- Logarithmic plotting of  $^{210}\text{Pb}_{\text{ex}}$  versus depth (Continued)

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