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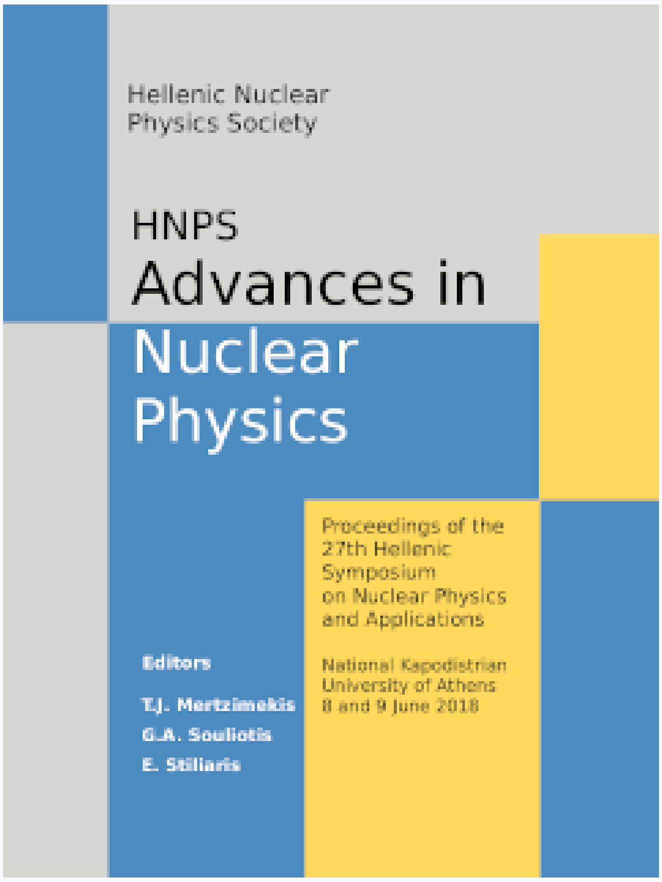

Application of radio-dating methods in marine areas of Greece

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Application of radio-dating methods in marine areas of Greece

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Abstract Chronology models based on radiotracers are useful for dating aquatic sediments influenced by industrialization. Among the many natural (e.g. ^{7}Be, ^{234}Th, ^{210}Pb) and artificial (e.g. ^{137}Cs, ^{238,239,240}Pu) radiotracers, the proper ones to study the industrial impact and reconstruct past events during the last 100 years, are ^{210}Pb and ^{137}Cs. Mining belongs to the industries with great ecological impact in the aquatic environment, as mines are situated near environmental sensitive areas such as coasts, rivers and lakes. Additionally, the accurate historical reconstruction may provide information regarding the mine footprint in the aquatic environment so as to assess the previous or the remaining contamination, in case of on-going or abandoned mines. In the present study, the retrospective investigation in a coastal area of Lavrio, near an abandoned metallic mine is attempted, using the ^{210}Pb and ^{137}Cs methodologies and the emerged difficulties are discussed.

Keywords radiotracers, ^{210}Pb radiochronology models, ^{137}Cs method, Lavrio

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INTRODUCTION

Contaminants such as radionuclides and heavy metals, tend to be trapped in aquatic environments and accumulated in sediments during processes such as adsorption and precipitation. These processes followed by sedimentation, contribute to the creation of a history repository regarding the temporal trend of contaminant input into aquatic environments. In the case where long-term monitoring data are scarce, the sedimentary records can provide retrospective information on the past events occurred in the aquatic area. According to [1], near marine areas well-established dating methods can be applied, based on two independent methodologies of ^{210}Pb (natural radionuclide) and ^{137}Cs (artificial radionuclide). However, many difficulties arise near coastal areas, such as the disturbed ocean floor due to physical processes (e.g. waves, tides) or anthropogenic activities (e.g. aquaculture, land use changes), which influence the ^{210}Pb profiles. Additionally, the low intensity signal of ^{137}Cs inputs hinders the application of ^{137}Cs methodology in many cases.

A sediment core was collected in the coastal area of Lavrio (Oxygono Bay) at Lavreotiki Peninsula, which was influenced by the long-term mining activities. Lavrio is located near an abandoned multi-metallic mine, has been developed on top of mine wastes and the marine coastal area received for many years disposed wastes. The main objectives of this work is to

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date the sediment core and to validate the dating with historical data. The applied methodology implemented two independent radio-dating methodologies (^{210}Pb, ^{137}Cs) and provided sedimentation rate data for Lavrio coastal area.

EXPERIMENTAL DETAILS

The experimental details regarding sampling, sample preparation and radionuclide measurements are described in detail elsewhere [2]. Briefly, a sediment core of approximately 50 cm depth was collected in Oxygono Bay and sliced per 2 cm. The activity concentrations of ^{137}Cs, ^{226}Ra and its progenies were obtained via gamma-ray spectrometry using the HPGe detector of Nuclear Laboratory of Physics Department of National Technical University of Athens (NTUA), while ^{210}Pb values were measured in the Department of Physics of the Aristotle University of Thessaloniki (AUTH).

Sedimentation models

The radio-chronological methodologies of ^{210}Pb ($t_{1/2}$=22.23 y) and ^{137}Cs ($t_{1/2}$=30.05 y) provide the temporal frame to interpret the records of sedimentation changes and when combined with historical data they offer retrospective studies beyond the time-scale of existing monitoring data [1]. Each approach is based on the fallout of excess ^{210}Pb ($^{210}Pb_{ex}$) and ^{137}Cs, and is characterized by specific assumptions and limitations such as that the sampling locations must have rapid sedimentation rates (SR > 0.03 cm y^{-1}), the geomorphology parameters (e.g. sediment type, bathymetry, current) allow the building of sedimentary records and the signal of ^{137}Cs is present in the area of study. The excess or unsupported ^{210}Pb near shore zones is an extra portion added in the existing ^{210}Pb which is in secular equilibrium with its parent nuclide (^{226}Ra). This extra portion can be supplied from atmospheric deposition, where ^{222}Rn (progeny of ^{226}Ra) exhales from the soils to the atmosphere, decays to ^{210}Pb, attaches to aerosols and deposits by wet and dry processes to the globe's surface [3]. The measured ^{210}Pb in the sediment is the sum of the supported ^{210}Pb (secular equilibrium with ^{226}Ra) and the unsupported (excess) ^{210}Pb, this by subtracting the ^{226}Ra activity concentration the excess portion is obtained.

Different ^{210}Pb models utilize the excess portion ($^{210}Pb_{ex}$) with the concentration in the sediment, the flux in the surface of the sediment and the mass accumulation rate in the surface sediment. All models assume that $^{210}Pb_{ex}$ decays as described by the radioactive decay law and there are no processes that redistribute $^{210}Pb_{ex}$. Nevertheless, the latter condition is not always met, thus the estimated dates by the models must be validated with independent observations. Usually, another independent dating method is applied (^{137}Cs), as well as historical data to verify the dating. The most used ^{210}Pb models are named Constant Initial Concentration model (CIC), Constant Rate of Supply model (CRS) and Constant Flux Constant Sedimentation model (CFCS) and as indicated by their names, each model is based on different assumptions. The equations describing briefly the ^{210}Pb (CIC, CRS, CFCS) and ^{137}Cs dating models are following [4]:

The Constant Initial Concentration model (CIC) assumes that the concentration of $^{210}Pb_{ex}$ is constant (C_0) when the layer i of the sediment core is formed and decays following the radioactive decay law. The estimated elapsed time applying the CIC model is given by:

$$t = \frac{1}{\lambda} \ln\left(\frac{C_0}{C_i}\right) \qquad \text{(Eq. 1)}$$

where, t is the elapsed time, λ is the disintegration constant of ^{210}Pb (0.03118 ± 0.00017 y^{-1}), C_0 is the $^{210}Pb_{ex}$ concentration in the first layer of the sediment core during the sampling date (t=0) and C_i is the concentration of layer i.

The constant Rate of Supply model (CRS) assumes that the flux of $^{210}Pb_{ex}$ is constant in the sediment surface and the age of i^{th} layer can be determined as:

$$t = \frac{1}{\lambda} \ln\left(\frac{A_0}{A_i}\right) \qquad \text{(Eq. 2)}$$

where A_0 is the inventory of $^{210}Pb_{ex}$ in the first layer of the sediment core during the sampling date (t=0) and A_i is the inventory of the i^{th} layer.

The fundamental assumption of the Constant Flux Constant Sedimentation model (CFCS) is the constant flux in the sediment surface and the constant mass accumulation rate of $^{210}Pb_{ex}$. The sedimentation rate (SR), and thus the elapsed time using this model, can be derived by the slope (λ/SR) of the equation:

$$\ln C_i = \ln C_0 - \frac{\lambda}{SR} m_i \qquad \text{(Eq. 3)}$$

where, SR is the sedimentation rate (cm y^{-1}) and m_i is the mass of depth i.

The dating model based on ^{137}Cs, associates the signals due to nuclear incidents of the aforementioned radionuclide with the depths of the ^{137}Cs peaks, so as to determine the sedimentation rate (SR):

$$SR = \frac{d_i}{t_0 - i} \qquad \text{(Eq. 4)}$$

where, d_i is the depths of ^{137}Cs peaks, t_0 is the sampling date (in y) and i are years of Chernobyl accident (i=1986) and nuclear tests (i=1963).

RESULTS AND DISCUSSION

All the dating models based either on ^{210}Pb (CIC, CRS, CFCS) or ^{137}Cs were applied in the sediment core collected in the coastal area of Lavrio in Oxygono Bay. The estimated elapsed times for the i^{th} depth of the sediment core are presented in Fig. 1.

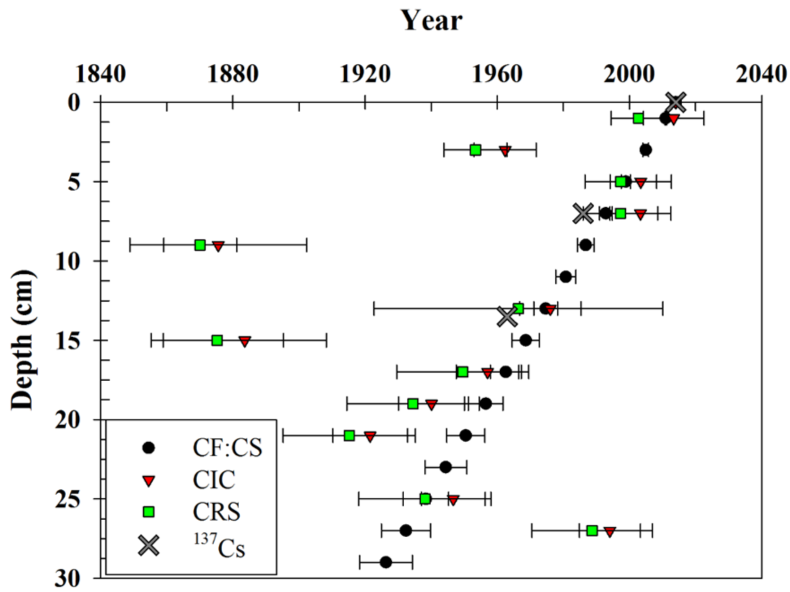
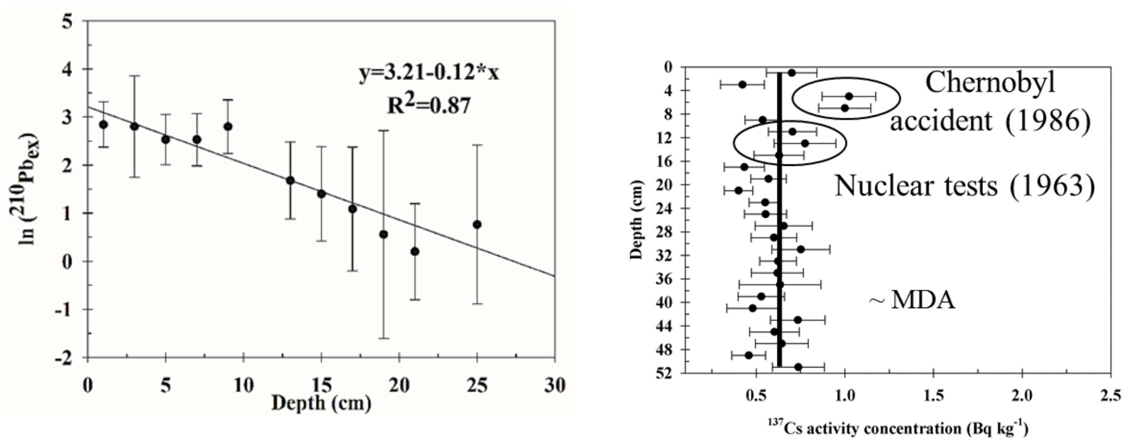


Fig. 1. The estimated elapsed time utilizing the ^{210}Pb (CIC, CRS, CFCS) and ^{137}Cs dating models. The ^{210}Pb models of Constant Initial Concentration model (CIC), Constant Rate of Supply model (CRS) and Constant Flux Constant Sedimentation model (CFCS) take into consideration different constant parameters of $^{210}Pb_{ex}$ deposition in the sediment surface such as concentration, flux, as well as mass accumulation rate and flux, respectively.

The CFCS model assumes a constant sedimentation rate and agrees –within uncertainties- with the sedimentation rate determination of ^{137}Cs model. However, the other ^{210}Pb models (CIC and CRS) assume a variable SR resulting in great time discrepancies along with depth or predicting newly formatted deep sediment layers, which is unnatural. Therefore, the CFCS and ^{137}Cs models were utilized for the estimation of the sedimentation rate in Oxygono Bay as shown in Fig. 2:



The slope of the logarithmized $^{210}Pb_{ex}$ concentration versus depth of sediment layers in the CFCS model can be utilized in the sedimentation rate (SR) estimation [5].

The ^{137}Cs peaks are correlated with the nuclear incidents. (MDA: Minimum Detectable Activity)

Fig. 2. The CFCS (left) and the ^{137}Cs (right) models implemented in the sediment core of Lavrio coast in Oxygono Bay.

The estimated sedimentation rate was found to be (0.27 ± 0.03) cm y^{-1} and (0.23 ± 0.01) cm y^{-1} using the ^{210}Pb and ^{137}Cs models, respectively. The mean sedimentation rate was utilized for the dating and the historical data of the Pb exploitation in Lavrio area were used for the validation of the radiochronology. The obtained dating by combining the two models resulted in a difference of 40 y from the historical data, thus the SR value of ^{137}Cs model was excluded as the low activity concentrations near the Minimum Detectable Activity (MDA) of the HPGe detector and the low statistics concealed the clear signal of ^{137}Cs as shown in Fig. 2. Thereinafter only the sedimentation rate of the CFCS model was applied for the sediment core dating and the difference from the historical data was found to be 30 y. This difference indicates that the main assumptions of the CFCS model do not meet. The ^{210}Pb models, assumes that the excess ^{210}Pb ($^{210}Pb_{ex}$) decays as implies the radioactive decay law, thus the logarithmized activity concentration of $^{210}Pb_{ex}$ must follow a linear decrease. However, some experimental points of the logarithmized $^{210}Pb_{ex}$ profile diverge from this assumption as shown in Fig. 2. By excluding these points the sedimentation rate using the CFCS model was found to be (0.33 ± 0.03) cm y^{-1} and the difference between the dating and the historical data was minimized to 10 y. This difference is within the time resolution of the method as the sediment core was sliced per 2 cm.

CONCLUSIONS

In the present study the radiodating models were tested and those with good agreement were utilized for the estimation of sedimentation rate in the coastal area of Lavrio in Oxygono Bay. Two independent radiodating methods based on ^{210}Pb (CFCS model) and ^{137}Cs were applied in the sediment core of Oxygono Bay and the dating was validated using the historical data of Pb exploitation. The discrepancies between the radiodating and the historical data, resulted in excluding the ^{137}Cs model and re-adjusting the CFCS model, as the fundamental assumptions of these models such as clear ^{137}Cs signals and $^{210}Pb_{ex}$ linear logarithmic decrease were not met. During this work a methodology was developed to optimize radiodating techniques in coastal areas characterized by low activity concentrations due to high sandy content.

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