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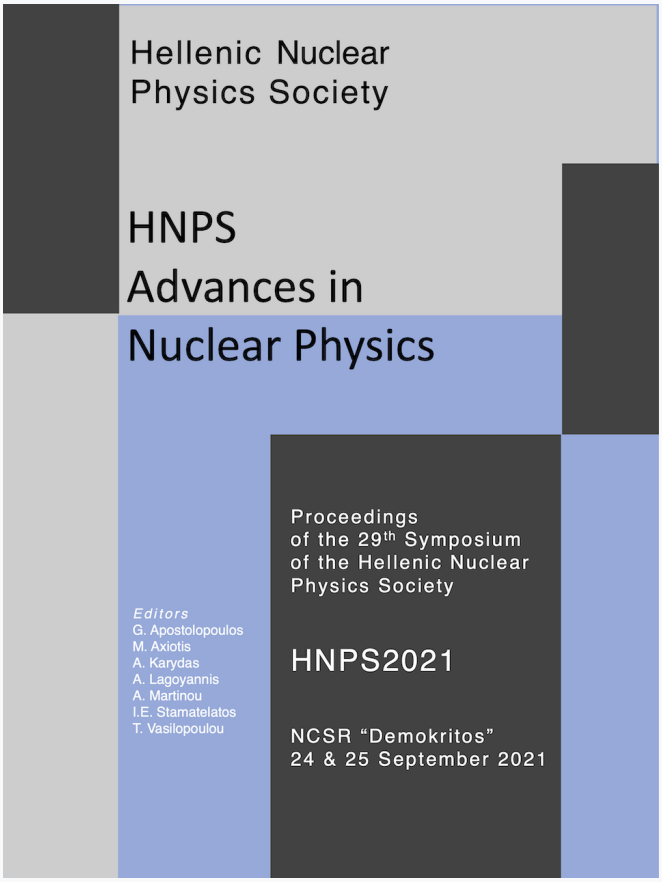

In Depth Analysis of a Sediment Core from North Aegean Sea

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In Depth Analysis of a Sediment Core from North Aegean Sea

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Abstract In this work a thorough analysis of the vertical distribution of natural and artificial radionuclides and trace elements in a sediment core sample was conducted. A sediment core of 39 cm length was grabbed from the deep-sea trench (1540 m depth) southern of Samothrace Island, Aegean Sea. The sediment core was then treated at the Hellenic Centre for Marine Research, separated at 1cm increment samples, ranging from 0cm to 39 cm, dried, and milled. A first analysis of the samples was also conducted at HCMR using gamma spectroscopic techniques. Further spectroscopic analysis continued at Nuclear Engineering Department of the National Technical University of Athens (NED-NTUA) using an Extended Range Germanium Detector. The following radionuclides were determined ^{210}Pb, ^{234}Th, ^{226}Ra, ^{228}Ra, ^{228}Th, ^{137}Cs and ^{40}K. Additionally, Instrumental Neutron Activation Analysis was used and the following major and trace elements were identified and quantified: Al, V, La, Sc, Mn, As, Ga, Cr, Zn, Co, Rb, Fe, K, Na and Ce. Based on the analysis of each sample, the vertical distribution of most natural radionuclides showed no significant variation. The vertical distribution of anthropogenic ^{137}Cs was very low and almost constant. Unsupported ^{210}Pb activity was about six times higher than ^{226}Ra, indicating high radon fluxes. Furthermore, neutron activation analysis results indicate that almost all elements are within the expected range based on literature, while K and Na show a slightly increased concentration. For the area, these findings indicate steady and calm sedimentation processes.

Keywords sediment core, γ-spectrometry, neutron activation, trace elements

INTRODUCTION

The research on radionuclide activity and trace element distribution in marine sediments is of great interest for the scientific community [1-4]. The vertical distribution of radionuclides and major and trace elements in marine sediment cores provides information regarding the sediment composition, sedimentation rate as well as, deposition and transfer processes. In this work, a sediment core of 39 cm length was grabbed from the deep-sea trench (1540 m depth) southern of Samothrace Island, Aegean Sea, by the HCMR's R/V AEGAEO using a box corer. The core was divided into 39 sediment samples, 1cm each. Each sample was dried to remove humidity and pulverized. An initial analysis of the samples was conducted at HCMR using gamma spectroscopic techniques and afterwards each sediment sample was analyzed at NED-NTUA by means of γ-spectrometry and Neutron Activation Analysis (NAA) to determine the vertical distribution of radionuclides and major and trace elements.

GAMMA-SPECTROMETRY AND NEUTRON ACTIVATION

Gamma-Spectrometry Analysis

All 39 sediment samples were analyzed using an Extended Range (XtRa) Germanium Detector with 104.5% relative efficiency and C-window coupled with a Compton Suppression System (CSS) [5]. Two cylindrical sample geometries of 18.10cm^3 and 8.72cm^3 were used for the analysis of the samples. Both geometries were calibrated via Monte Carlo simulation and were experimentally

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validated. Selected samples, were sealed for radon in order to establish equilibrium between ^{226}Ra and its short lived progenies [6]. This way, determination of activity concentration for both total and unsupported ^{210}Pb was possible, by subtracting ^{226}Ra activity from $^{210}Pb_{total}$.

Neutron Activation Analysis

Instrumental Neutron Activation Analysis (INAA) is used by NED-NTUA to identify and determine the major and trace element content of various types of samples [7]. In this work, selected increments of the sediment core sample underwent neutron activation using a ^{241}Am-Be source of ≈10Ci (neutron flux $2.2 \cdot 10^7$ n/s ± 10%). Two irradiation scenarios were applied. The first irradiation scenario requires: 1 hour of irradiation, 1 min delay and 15 mins of γ-spectrometric analysis. The second scenario requires: 15 days irradiation, 5 hours delay and γ-spectrometric analysis for 1 and 5 days consecutively. For the quantitative calibration of the whole process the IAEA Standard Reference Material 2702 "Inorganics in Marine Sediment" was used.

RESULTS AND DISCUSSION

The radionuclides detected via γ-spectrometric analysis were: ^{210}Pb, ^{234}Th, ^{226}Ra, ^{228}Ra, ^{228}Th, ^{137}Cs and ^{40}K [2,8-9]. Since significant disruption of radioactive equilibrium between ^{210}Pb and ^{226}Ra was observed, the measured $^{210}Pb_{total}$ was divided into $^{210}Pb_{supported}$ by ^{226}Ra and $^{210}Pb_{unsupported}$. Unsupported ^{210}Pb activity was about six times higher than ^{226}Ra. The origin of the unsupported ^{210}Pb is related with radon gas diffusive transportation in the sediment and emanation as well as radon progenies attached on aerosols which fall in the seawater due to precipitation. The vertical distribution of ^{137}Cs, $^{210}Pb_{total}$, ^{226}Ra and $^{210}Pb_{unsupported}$ are shown in Figures 1-4, with the error bars corresponding to combined standard uncertainties. The vertical distribution of the majority of the other natural radionuclides showed no significant variation or trend and ranged: for ^{234}Th between 34 and 54 Bq/kg, for ^{228}Ra between 62 and 77 Bq/kg, for ^{228}Th between 50 and 55 Bq/kg and for ^{40}K between 604 and 758 Bq/kg,. The mean activity ratio of ^{226}Ra/^{234}Th was estimated to 0.94 ± 0.03, indicating radioactive equilibrium between them, while the mean activity ratio of ^{228}Ra/^{234}Th was estimated to 1.60 ± 0.05 which is slightly higher than the typical range of 0.8-1.4 reported for ^{232}Th/^{238}U ratio for soil [6]. According to the $^{210}Pb_{unsupported}$ distribution and the almost constant activity concentrations of the other radionuclides, a continuous undisturbed sedimentation process in the location is evident. The vertical distribution of the anthropogenic ^{137}Cs was very low and no peak related to nuclear incidents was observed. However, the presence of ^{137}Cs in sediment depth down to 35 cm indicates high mobility of cesium in the sediment matrix. The molecular mobility of cesium is mainly attributed to diffusive transportation downwards the sediment column through pore water [2].

Table 1. Vertical distribution in ppm of Mn, As, Cr, Zn, Co, Rb and Fe

Depth (cm)	Mn (ppm)	As (ppm)	Cr (ppm)	Zn (ppm)	Co (ppm)	Rb (ppm)	Fe (ppm)
1-2	3013 ± 83	24 ± 6	164 ± 48	428 ± 316	28 ± 13	150 ± 90	39086 ± 3295
5-6	3447 ± 93	23 ± 5	139 ± 46	ND	31 ± 13	129 ± 77	43697 ± 3080
9-10	3504 ± 94	20 ± 5	156 ± 45	387 ± 258	22 ± 10	104 ± 68	44355 ± 2901
17-18	3509 ± 95	19 ± 5	120 ± 37	ND	23 ± 11	104 ± 68	44222 ± 2914
23-24	3051 ± 83	21 ± 6	114 ± 63	702 ± 424	ND	ND	39832 ± 4769
27-28	2968 ± 80	18 ± 5	96 ± 43	502 ± 292	17 ± 9	105 ± 66	42722 ± 2794
33-34	2981 ± 81	14 ± 4	126 ± 40	ND	22 ± 10	102 ± 66	41378 ± 2789
38-39	2461 ± 67	22 ± 5	129 ± 40	ND	23 ± 10	101 ± 66	40666 ± 2845

Table 2. Vertical distribution in ppm of K, Na, Ga, Ce, Sc and La.

Depth (cm)	K (ppm)	Na (ppm)	Ga (ppm)	Ce (ppm)	Sc (ppm)	La (ppm)
1-2	23073 ± 971	17809 ± 283	18 ± 4	ND	18 ± 1	33 ± 6
5-6	24062 ± 954	18138 ± 284	20 ± 5	88 ± 20	18 ± 1	32 ± 5
9-10	23378 ± 909	19040 ± 300	16 ± 3	71 ± 18	17 ± 1	30 ± 4
17-18	22968 ± 911	17259 ± 274	18 ± 4	76 ± 18	17 ± 1	33 ± 5
23-24	22757 ± 877	16910 ± 267	8 ± 4	ND	16 ± 2	31 ± 6
27-28	22695 ± 883	17366 ± 276	14 ± 3	ND	17 ± 1	30 ± 4
33-34	23327 ± 890	17872 ± 290	14 ± 3	ND	16 ± 1	32 ± 4
38-39	21776 ± 847	18151 ± 284	16 ± 4	72 ± 18	16 ± 1	29 ± 4

A total of 15 major and trace elements, among which 3 rare earth elements, were identified and quantified by Neutron Activation Analysis in selected samples [1]. Specifically Al and V, which were identified by the first irradiation and analysis scenario, showed uniformity in their depth distribution and ranged from 64500-73000ppm and 107-183ppm respectively. The vertical distribution of the rest of the detected elements, which were identified from the second irradiation scenario are shown in Tables 1 and 2.

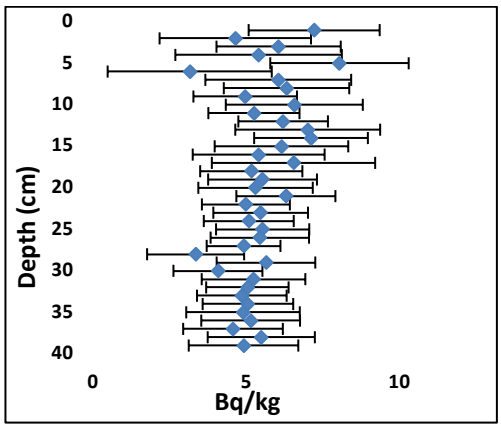

Fig. 1. *Depth distribution of ^{137}Cs*

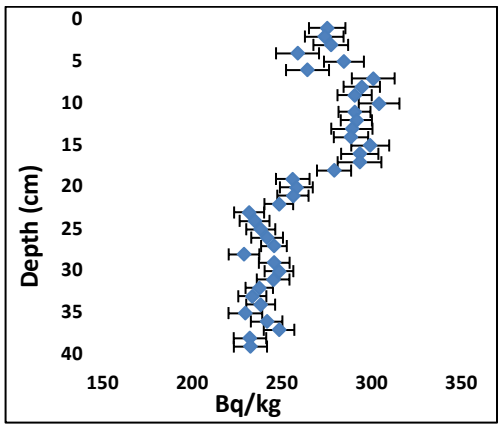

Fig. 2. *Depth distribution of $^{210}Pb_{total}$*

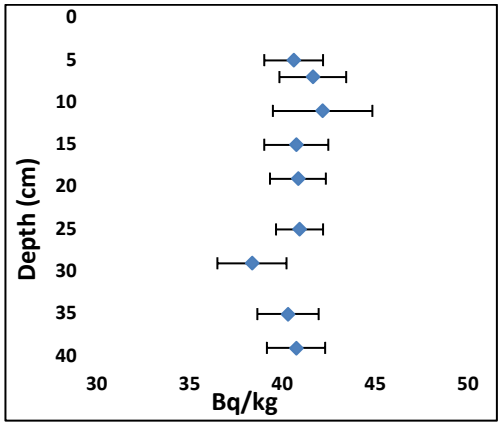

Fig. 3. *Depth distribution of ^{226}Ra*

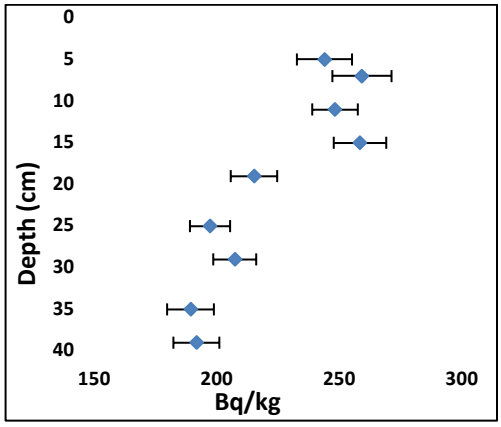

Fig. 4. *Depth distribution of $^{210}Pb_{unsupported}$*

It is worth mentioning that there was strong indication of Samarium (Sm) in the samples, however, quantification was not possible due to strong Uranium interference [10].

CONCLUSIONS

The vertical distribution of most natural radionuclides showed no significant variation indicating continuous, undisturbed sedimentation process in the studied location. The mean activity ratio of $^{226}Ra/^{234}Th$ indicates radioactive equilibrium and the mean activity ratio of $^{228}Ra/^{234}Th$ was slightly higher than the typical range of 0.8 – 1.4 [11]. For the area, both findings indicate steady and smooth sedimentation processes. The vertical distribution of anthropogenic ^{137}Cs was very low and almost constant. No peak related to nuclear incidents was observed indicating very low sedimentation rate which is expected for deep sea. After deposition at the seafloor, ^{137}Cs diffuses through the sediment pores obtaining almost uniform vertical distribution. Unsupported ^{210}Pb activity was about six times higher than ^{226}Ra, indicating that radon emanation as well as atmospheric deposition of radon progenies have lead to activity levels of the upper (1-15cm) sediment layers about 20% higher than the activity of the deeper layers. Trace and major elements obtained by NAA are within the expected range based on literature [12], while K and Na show a slightly increased concentration. In any case, no vertical profile was observed.

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